

Review on Synthesis, Characterizations, and Electrochemical Properties of Cathode Materials for Lithium Ion Batteries

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Abstract

The development of cutting-edge cathode materials is a challenging research topic aiming to improve the energy and power densities of lithium ion batteries (LIB) to cover the increasing demands for energy storage devices. Therefore, highly needed further improvements in the performance characteristics of Li-ion batteries are largely dependent on our ability to develop novel materials with greatly improved Li ion storage capacities. Three different types of cathode materials including intercalation, alloying and conversion materials are reviewed in this paper in order to orientate our researches towards highly performant LIBs batteries. This includes characteristics of different cathode materials and approaches for improving their performances.

Keywords: Lithium ion battery; Cathode materials; Insertion; Conversion; Specific capacity

Introduction

Since their commercialization by SONY in 1991 [1], lithium ion batteries (LIBs) have made significant progress in terms of safety, electrochemical properties such as capacity, power, and cycling stability and it has the highest energy density comparing to other secondary batteries such as nickel-cadmium and nickel-metal hydride. A great part of such progress can be attributed to the introduction of new materials with higher capacity, higher potential, and enhanced thermal stability. Along this journey, numerous studies have been conducted in order to find cathode materials for LIBs with higher capacity to allow for practical applications in plug-in hybrid electric vehicles, large-scale power generation systems, and critical space and aeronautical applications.

In this paper, a comprehensive literature review of cathode materials used or proposed for LIBs is attempted. Since the focus is in this paper on capacity, three areas will be highlighted through the review: synthesis methods, characterization techniques, and electrochemical properties related to capacity, namely initial discharge capacity and cyclability. Out of several possible categorization schemes of cathode materials for LIBs, a structure-based scheme (layered, spinel, olivine, etc.) seems to be dominant as used by most reviewers such as Whittingham [2] and Xu et al. [3]. Also other kinds of cathodes have been studied (air, sulfur, organics and conversion like transition metal FeF_3).

Different synthesis techniques were developed and utilized to produce cathode materials studied including solid-state reaction, sol-gel method, hydrothermal synthesis, and co-precipitation, emulsion-drying, spray pyrolysis, and many others (Figure 1 and Table 1). The electrochemical properties of materials formed, such as initial discharge, cyclability, and capacity retention rate, as well as the morphological properties such as surface texture, grain size, size distribution, and crystallinity, are directly affected by the synthesis method (Table 1). Spray pyrolysis method, in particular, allows for controlling the particle size distribution which provides a powerful tool to control the chemical structural and morphology properties of a material.

Structural characterization of cathode materials synthesized have been done by using different techniques such as X-ray diffraction (XRD), transmission electron microscopy (TEM), scanning electron microscopy (SEM), differential scanning calorimetry (DSC), dynamic light scattering (DLS), and Raman spectroscopy. These techniques have been utilized to determine crystal structures, chemical and

phase compositions, surface morphological characteristics, and microstructural features of synthesized materials.

Although LIB's have many advantages, it has its drawbacks since it needs high protection circuit to keep both current and voltage within

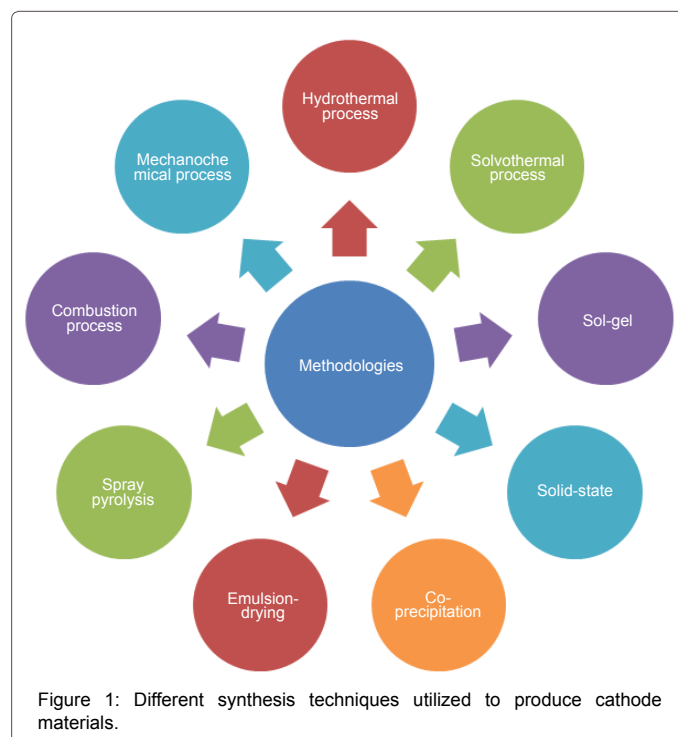


Figure 1: Different synthesis techniques utilized to produce cathode materials.

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Material	Research group (year)	Synthesis method	Characterization technique	Initial capacity (mAhg ⁻¹)	Number of cycles: Capacity loss
Layered metal oxides					
Li _x CoO ₂ (0 ≤ x ≤ 1)	Mizushima et al. [6]	Electrochemical extraction from the LiCoO ₂ rocksalt structure	XRD	-	-
HT-LiCoO ₂	Shlyakhtin et al. [18]	Freeze-drying method	XRD; SEM; TEM; SAED	135-138	10: 10-14%
LiCoO ₂	Gan et al. [14]	Carbon combustion synthesis	XRD; SEM	148	10: 3%
Carbon-coated LiCoO ₂	Cao et al. [25]	Commercial LiCoO ₂ powder with milling for 24 h at 300 rpm	XRD; SEM; TEM; EIS	130 at 0.1°C	-
Carbon-coated LiCoO ₂ thin film with PVDF-HFP gel electrolyte	Park et al. [11]	Sol-gel process and Screen printing	XRD; SEM	110	-
ZnO-coated LiCoO ₂	Chang et al. [36]	Plasma-enhanced chemical vapour deposition	XRD; SEM; AAS; TGA; DSC; BET; EIS	~ 178 at 1°C	30: ~ 33%
Al ₂ O ₃ -coated LiCoO ₂	Lu et al. [12]	<i>In situ</i> sol-gel process followed by calcination at 1123 K for 12 h	XRD; SEM; TEM; XPS; DSC	195 (1.0 wt% Al ₂ O ₃)	30: 15%
Li ₄ Ti ₅ O ₁₂ -coated LiCoO ₂	Yi et al. [32]	Commercial LiCoO ₂ powder and coating with sol-gel process	XRD; SEM	142.7 (3 wt% Li ₄ Ti ₅ O ₁₂)	40: 2.7%
FePO ₄ -coated LiCoO ₂	Li et al. [16]	Co-precipitation method followed by a high-temperature treatment	XRD; SEM; TEM; EDS; EIS; AES	146 (3 wt% FePO ₄)	400: 11.3%
High-density spherical LiCoO ₂	Ying et al. [24]	Controlled crystallization method	XRD; SEM	148.4 at 0.2°C	40: 2.4%
ZnO-coated LiCoO ₂	Fang et al. [39]	Commercial LiCoO ₂ powders coated with wet chemical method	XRD; SEM; ICP-AES	~ 188 at 0.1°C (0.2 wt% ZnO)	30: 10.4%
ZnO-coated LiCoO ₂	Fang and Duh [38]	Wet chemical method with a calcination process	XRD; SEM	~ 185 (calcined at 650°C)	30: ~ 13.5%
SrO/Li ₂ O/La ₂ O ₃ /Ta ₂ O ₅ /TiO ₂ -coated LiCoO ₂	Wang et al. [34]	Commercial LiCoO ₂ powders coated with Sol-gel method	XRD; SEM	~ 140 at 1°C (1.0 wt% SrO/Li ₂ O/La ₂ O ₃ /Ta ₂ O ₅ /TiO ₂)	900: ~ 25%
M-coated LiCoO ₂ (M=TiO ₂ ZrO ₂ or ZrTiO ₄)	Fey et al. [35]	Sol-gel and mechano-thermal processes	XRD; TEM	167 (ZrO ₂ -coated) 165 (TiO ₂ -coated) 168 (ZrTiO ₄ -coated)	110: 20% 24: 20% 37: 20%
Li ₂ PO ₂ N-coated LiCoO ₂	Choi et al. [31]	RF magnetron sputtering method	EDS; SEM; DSC; EIS	~ 185 at 0.2°C	50: ~ 16%
LiCoO ₂ /LiNi _{0.8} Co _{0.17} Al _{0.03} O ₂	Lin et al. [236]	Commercial products	-	186 (for pure LiNi _{0.8} Co _{0.17} Al _{0.03} O ₂) 161.4 (for 50:50 mixture)	20: 5%
LiCoO ₂ thin film	Park et al. [237]	Screen-printing method using ethyl-cellulose-based paste	SEM	133	-
LiCoO ₂ /polyacrylonitrile/binary conductive additives composite	Zhang et al. [238]	Commercial products	XRD; SEM; TEM	155.8 at 0.5°C (~ 12.5 wt% binary conductive additives)	-
poly(3,4-dioxyethylenethiophene)/LiCoO ₂ composite	Her et al. [239]	Electrochemical deposition process	DSC; SEM; EDS	-	50: ~ 0%
Ag-doped LiCoO ₂	Huang et al. [28]	Commercial LiCoO ₂ powder with milling for 4h in alcohol	XRD; SEM	172.3 at 1°C 133.1 at 10°C	50: 22.29% at 1°C 50: 4.88% at 10°C
Li _x MnO ₂ (0.4 ≤ x ≤ 0.7)	Pistoia et al. [240]	Solid-state reaction	XRD, TGA, TDA	~ 108 (x=0.55)	65: ~ 16%
LiNiO ₂	Yamada et al. [241]	Solid-state reaction at 500-900°C for 5 h	XRD	197 at 1°C (at 700°C in O ₂) 173 at 1°C (at 700°C in air)	12: ~ 0%
LiNiO ₂	Kalyani et al. [21]	Microwave synthesis with O ₂ heat pre-treatment	XRD; BET; SEM	163	<5%
Co-coated LiNiO ₂	Sheng-wen et al. [242]	Co-precipitation method	XRD; SEM; TEM; XPS	180	20: 0.07%/cycle
Ga-doped LiNiO ₂	Nishida et al. [243]	Sol-gel method	XRD	190	100: 5%
LiNi _{1-x} Mg _x O ₂ (x=0, 0.1, 0.2, 0.3, or 0.5)	Sathiyamoorthi et al. [244]	Solid-state reaction at 600°C for 8 h	XRD; FTIR; SEM; EDS; TEM	190 (x=0.2)	25: 2.4%
LiCo _{1-x} Ni _x O ₂ (0 ≤ x ≤ 0.2)	Gummow and Thackeray [245]	Solid-state reaction at 400°C for 48-120 h	XRD	-	-
LiNi _{0.5} Mn _{0.5} O ₂	Abdel-Ghany et al. [246]	Wet-chemical method	XRD; SEM	166	30: 5% at 900°C

$\text{LiNi}_{0.9}\text{Co}_{0.1}\text{O}_2$	Shi et al. [247]	Rheological phase reaction method	XRD; SEM	199 (synthesized at 800°C)	15: 27.1%
$\text{LiNi}_{0.8}\text{Co}_{0.2}\text{O}_2$ powder	Sivaprakash et al. [248]	Solid-state reaction at 700-800°C for 15-48 h with intermittent grinding	XRD	~ 157	20:~11%
$\text{LiMn}_x\text{Co}_y\text{Ni}_{1-x-y}\text{O}_2$	Wang et al. [249]	Solid-state reaction at 800-900°C	XRD; SEM	180 (x=0.2 and y=0.25)	50: 15%
$\text{LiCo}_{1/3}\text{Ni}_{1/3}\text{Mn}_{1/3}\text{O}_2$ powder	Ogihara et al. [250]	Spray pyrolysis	SEM; XRD; BET; AAS	170	500: 20%
$\text{LiNi}_{0.7}\text{Co}_{0.2}\text{Ti}_{0.05}\text{M}_{0.05}\text{O}_2$ (M=Mg, Al or Zn)	Subramanian and Fey [41]	Solid-state reaction at 800°C for 12 h	XRD; SEM; ICP-AES	149	30: -2% (Mg-doped)
$\text{Li}_{1+x}\text{Ni}_{0.75}\text{Co}_{0.25}\text{Mg}_x\text{O}_{2(1+x)}$	Chang et al. [251]	Sol-gel method	XRD; TGA; TDA	~170 (x=0.1)	30: ~ 12%
$\text{LiAl}_x\text{Co}_{1-x}\text{O}_2$ (0 ≤ x ≤ 0.3)	Myung et al. [26]	Emulsion-drying method	XRD; TEM	110 (x=0.1)	-
$\text{LiAl}_x\text{Co}_{1-x}\text{O}_2$ (0.1 ≤ x ≤ 0.3)	Huang et al. [252]	Solid-state reaction 550°C for 20 h and at 750°C for 24 h with intermittent grinding	XRD	153 (x=0.1)	15% at x=0.1
$\text{LiAl}_{0.05}\text{Ni}_{0.95-x}\text{Co}_x\text{O}_2$ (x=0.1 or 0.15)	Zhu et al. [253]	Solid-state reaction at 725°C for 24 h	XRD; SEM; TG-TDA	186.2 (x=0.1)	10: 3.2%
$\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Al}_{1/3-x}\text{Co}_x\text{O}_2$ (0 ≤ x ≤ 1/3)	Ren et al. [254]	Solid-state reaction at 900°C for 20h	XRD; SEM	~170 at x: 1/3	20:~0
$0.5\text{Li}_2\text{MnO}_3\text{-LiNi}_{0.5}\text{Mn}_{0.5}\text{O}_2$	Zhong et al. [255]	Low temperature combustion followed by annealing treatment	XRD; SEM	139 at 0.1°C	50: -44%
Macroporous $\text{LiNi}_{0.5}\text{Mn}_{0.5}\text{O}_2$	Wang et al. [256]	Solid-state reaction method at 900°C for 10 h	XRD; SEM; BET; EIS	174 at 1°C	10: <5%
Al_2O_3 -coated $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$	Kim et al. [257]	Co-precipitation method at 1000°C for 10 h	XRD; SEM; EIS	173.8	50: 10.9%
Carbon-coated $\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$	Kim et al. [258]	Co-precipitation at 350°C for 3 h	XRD; DSC; TGA; SEM; TEM	150 at 1 wt% coating	50: 2% at 1 wt% coating
nano-crystalline LiCoO_2 -coated $\text{Li}_{1.05}\text{Ni}_{0.35}\text{Co}_{0.25}\text{Mn}_{0.4}\text{O}_2$	Son and Cairns [259]	Sol-gel method	XRD; TEM	164.1 at 0.1°C 151.2 at 0.2°C	20: ~ 0% at 0.1°C 20: 7.8% at 0.2°C
Spherical $\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$	Zhi-min, et al. [260]	Spray-drying method	XRD; SEM	153 at 0.2°C	40: ~ 6%
$\text{Li}_{1+x}(\text{Ni}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3})_{1-x}\text{O}_2$	Zhang et al. [261]	Co-precipitation method	XRD; SEM; ICP	150 at 1°C	30: 5%
La_2O_3 -coated $\text{LiNi}_{0.8}\text{Co}_{0.2}\text{O}_2$	Fey et al. [262]		XRD; SEM; TEM; XPS; BET		
$\text{LiNi}_{0.67}\text{Co}_{0.15}\text{Mn}_{0.18}\text{O}_2$	Sun et al. [263]	Co-precipitation at 750°C for 20 h and at 850°C for 15h	XRD; EPMA; SEM	207 at 55°C	50: ~ 7% at 55°C
Spinel lithium metal oxides					
LiMn_2O_4	Pistoia and Rosati [264]	Solid-state reaction at 730°C for 6 h	SEM	~ 120	80: ~ 16%
LiMn_2O_4	Li and Xu	Solid-state reaction at 600°C for 6 h and 750°C for 72 h with intermittent grinding		112	200: 6.50%
Carbon/ $\text{Li}_{1+x}\text{Mn}_2\text{O}_4$ (0.9 ≤ x ≤ 1.2)	Guyomard and Tarascon [45]	Solid-state reaction at 800°C for 24 h	TGA	~ 125	200: ~ 12%
$\text{Li}_{1+x}\text{M}_y\text{Mn}_{2-(x+y)}\text{O}_{4+2}$ (M= Li, Cu, Zn, Ni, Co, Fe, Cr, Ga, Al, B, or Ti)	Pistoia et al. [240]	Solid-state reaction at 730°C for 72 h		~ 100	150: ~ 7%
$\text{Li}_2\text{Cr}_x\text{Mn}_{2-x}\text{O}_4$ (0 ≤ x ≤ 2)	Davidson et al. [76]	Solid-state reaction at 600°C for 3 h and at 1000°C for 72 h	XRD	~ 32 at 0.05°C (x=0.5)	25: ~ 28%
$\text{LiCr}_x\text{Mn}_{2-x}\text{O}_4$ (0 ≤ x ≤ 0.1)	Zhang et al. [52]	Solid-state reaction at 600°C for 15 h and 650°C for 48 h with intermittent grinding followed by slow cooling	TGA; XRD; BET	118 (x=0.1)	200: 6.70%
$\text{LiCo}_x\text{Mn}_{2-x}\text{O}_4$ (0 ≤ x ≤ 0.33)	Arora et al. [58]	Solid-state reaction preheating at 600°C for 6 h and heating 750°C for 72 h with intermittent grinding followed by slow cooling	XRD; SEM; BET	100	85: ~ 3%
$\text{LiCr}_x\text{Mn}_{2-x}\text{O}_4$ (x=0), 0.04, 0.06 or 0.1)	Wang et al. [75]	Pechini method	XRD; neutron diffraction; EDS	122(x=0.04)	50: 7%

$\text{LiMn}_{0.05}\text{Mn}_{1.95}\text{O}_4$ (M=Li, Al, Co, Ni, or B)	Lee et al. [74]	Citrate gel method	XRD	125 (Ni-doped) 122 (Co-doped)	110: ~ 2% 110: ~ 4%
$\text{LiAl}_{0.24}\text{Mn}_{1.76}\text{O}_{3.98}\text{S}_{0.02}$	Sun et al. [265]	Sol-gel method	XRD; TEM	104 at 50°C 98.5 at 80°C	70: 2.4% at 50°C 70: 8.4% at 80°C
$\text{LiAl}_x\text{Mn}_{2-x}\text{O}_4$ ($0 \leq x \leq 0.6$)	Myung et al. [26]	Emulsion-drying method		114(x=0.2)	50: 4% at 45°C
$\text{LiAl}_x\text{Mn}_{2-x}\text{O}_{4.5}$ (x=0.1, 0.2, 0.3, or 0.4) $0.05 \leq \delta \leq 0.07$	Cho et al. [266]	Solid-state reaction at 750°C for 72 h with intermittent grinding	XRD; DSC	125 (x=0.1 and $\delta=0.05$) 116 (x=0.2 and $\delta=0.07$)	50: ~ 5% 50: ~ 3%
$\text{LiAl}_x\text{Mn}_{2-x}\text{O}_{4-z}\text{F}_z$ (x=0.1 or 0.2; $0 \leq z \leq 0.5$)	Amatucci et al. [177,187]	Solid-state reaction at 800°C for 24 h		~100 (x=0.2; z=0.5)	400: 15% at 55°C
$\text{LiAl}_{0.18}\text{Mn}_{1.82}\text{O}_{3.97}\text{S}_{0.03}$	Sun et al. [267]	Sol-gel method	XRD; SEM	107 at 25 and 50°C 100 at 80°C	50: 3% at 25 and 50°C
$\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_{4-x}\text{S}_x$ (x=0 or 0.05)	Sun et al. [268]	Co-precipitation at 500°C and 800°C	XRD; SEM; TEM	122 (x=0.05 at 500°C)	50: 4%
$\text{LiMn}_{1.5}\text{Ni}_{0.5-x}\text{Co}_x\text{O}_4$ ($0 \leq x \leq 0.5$)	Wu et al. [269]	Spray-drying method	XRD; SEM	112 (x=0.2)	20: 1.80%
$\text{LiAl}_{0.05}\text{Mn}_{1.95}\text{O}_4$	Yi et al. [273]	Adipic acid-assisted sol-gel method at 800°C		128 (D50=17.3 pm)	50: 8.30%
$\text{LiGa}_x\text{Mn}_{2-x}\text{O}_4$ ($0 \leq x \leq 0.05$)	Liu et al. [276]	Sol-gel method		118 (x=0.05)	50: 9%
$\text{Ag}_x/\text{LiMn}_2\text{O}_4$ ($0 \leq x \leq 0.3$)	Zhou et al. [77]	Thermal decomposition		95 (x=0.2)	40: 3.30%
$\text{LiNi}_{0.4}\text{Mn}_{1.6}\text{O}_4$	Patoux et al.	Solid-state reaction at 600-900°C for 10 h		129	1000: 20%
$\text{LiMn}_{1.8}\text{Li}_{0.1}\text{Ni}_{0.1}\text{O}_{4-n}\text{F}_n$ (n=0, 0.1 or 0.15)	Luo et al.	Solid-state reaction at 450°C for 12 h		80 (n=0)	50: ~ 1%
$\text{LiMn}_x\text{Mn}_{2-x}\text{O}_{4-y}\text{Br}_y$ ($0 \leq x \leq 0.15$; $0 \leq y \leq 0.05$)	Huang et al.	Solid-state coordination at 700°C for 10 h		104 (x=0.15; y=0.05)	100: 14.50%
$\text{LiMn}_{1.5}\text{Ni}_{0.42}\text{Ga}_{0.08}\text{O}_4$	Shin and Manthiram [270]	Hydroxide precursor method	XRD; EIS; FTIR; TOF-SIMS	124 at 25°C 121 at 55°C	100: ~ 1%
$\text{Li}_{1.15}\text{Mn}_{1.96}\text{Co}_{0.03}\text{Gd}_{0.01}\text{O}_{4+5}$	Sun et al. [271]	Solid-state reaction at 850°C for 5 h and 600°C for 10 h	XRD; SEM	128.1 at 25°C 126.5 at 50°C	100: 0.017%/cycle at 25°C 100: 0.098%/cycle at 50°C
$\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ powder	Ogihara et al. [250]	Spray pyrolysis	SEM; XRD; BET; AAS	130	500: 8%
$\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ particles	Yang et al. [272]	Urea combustion method	XRD; FTIR; SEM	133.6	20: 0.4%
$\text{LiLa}_{0.01}\text{Mn}_{1.99}\text{O}_{3.99}\text{F}_{0.01}$	Yi et al. [273]	Ultrasonic-assisted sol-gel method	XRD; TGA; TG-TDA; SEM	126	50: 92.1%
$\text{Li}_{1.05}\text{Mn}_{0.02}\text{Mn}_{1.98}\text{O}_{3.98}\text{N}_{0.02}$ (M=Ga ³⁺ , Al ³⁺ or Co ³⁺ ; N=S ²⁻ or F ⁻)	Amaral et al. [274]	Solid-state reaction at 750°C for 72 h	XRD; SEM	120 (M=Ga and N=S ²⁻)	300: 4% (M=Ga and N=S ²⁻)
$\text{LiCr}_x\text{Mn}_{2-x}\text{O}_4$	Song et al. [275]	Solid-state reaction at 450-750°C for 2 h	XRD; SEM; ICP-AES	120 (synthesized at 650°C)	100: 13%
LiFePO ₄ -coated $\text{LiMn}_{1.5}\text{Ni}_{0.5}\text{O}_4$	Liu et al. [276]	Sol-gel method using citric acid	XRD; SEM; TEM; EDS	~ 110 at 1°C	140: 25% at 1°C
Gold-coated LiMn_2O_4	Tu et al. [277]	Solid-state reaction at 750°C for 20 h and coating by ion sputtering method	XRD; EIS; EDS	~ 126	50: ~ 6.3%
Li ₃ PO ₄ -coated LiMn_2O_4	Li et al. [67]	sol-gel method and coating dry ball-milling method	XRD; SEM; EIS	112.4 at 55°C	100: 15% at 55°C
ZnO-coated $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ powder	Sun et al. [278]	Sol-gel method and coating by in situ mixing for 4 h	XRD; TGA; EDS; SEM; TEM	137 at 55°C	50: ~ 0 at 55°C
Co-doped $\text{LiCo}_y\text{Mn}_{2-y}\text{O}_4$ (y=0.05-0.33)	Arora et al. [58]	Solid-state reaction at 600°C for 6 h and 750°C for 72 h	XRD; SEM; BET	~ 105 (y=0.16)	25: ~ 3%
$\text{LiMn}_2\text{O}_4/\text{C}$	Huang and Bruce [279]	Modified sol-gel method	XRD	150 at 0.5°C (synthesized at 200°C)	300: 40%
LiMn_2O_4	Santiago et al. [47]	Combustion of manganese nitrate tetrahydrate and urea	XRD; EIS	107	-
LiMn_2O_4	Yi et al. [46]	Adipic acid-assisted sol-gel method a	XRD; TGA; TG-DTA; XRD; XPS; SEM; ICP-MS	90.7 (synthesized at 350°C) 130.1 (synthesized at 800°C)	50: 6.4% 50: 14.8%
$\text{LiZn}_x\text{Mn}_{2-x}\text{O}_4$ ($0 \leq x \leq 0.15$)	Arumugam et al. [55]	Sol-gel method using aqueous solutions of metal nitrates and succinic acid	XRD; SEM; TEM	137 at 0.5°C (x=0) 122 at 0.5°C (x=0.10)	100: 41% 100: 13%

Olivine transition metal phosphates and silicates					
Mesoporous LiFePO ₄	Ren and Bruce [280]	Solid-state reaction at 550°C for 5 h	XRD; TEM; BET	155 at 0.1°C 127 at 1°C	30: ~ 0%
LiFePO ₄ nanorods	Changa et al. [281]	Hydrothermal synthesis	XRD; SEM; TEM; Mössbauerspectroscopy	~ 144 at 0.5°C	20: ~ 8%
LiFePO ₄ /graphene/C composite	Su et al. [282]	<i>In situ</i> solvothermal method	XRD; SEM; TEM	163.7 at 0.1°C 114 at 5°C	30: 3% at 0.1°C (LiFePO ₄ /graphene2%/C6%)
nanoporous LiFePO ₄ /C composite	Su et al. [283]	Hydrothermal synthesis	XRD; SEM; TEM	155.3	50: 3%
nanoporous LiFePO ₄ /C composite	Yang and Gao [284]	Impregnation from ethanol solution	XRD; TGA; DSC; TEM	68 at 50°C	50: 12%
LiFePO ₄ /C	Yang et al. [83]	Ultrasonic spray pyrolysis	XRD; TEM; SEM	150 at 1°C and 50°C (15 wt% C)	30: 14.5%
LiFePO ₄ -C/solid polymer electrolyte	Jin et al. [285]	Hydrothermal synthesis at 150°C	XRD; TEM	128 (5 wt% C)	30: 0.78%
C-LiFePO ₄ /polypyrrole composite	Boyano et al. [286]	<i>In situ</i> electrodeposition method	XRD; SEM; TGA	154 at 0.1°C (20% PPy)	120: ~ 0%
LiFePO ₄ /C composite fiber	Toprakci et al. [287]	Combination of electrospinning and sol-gel methods	XRD; SEM; TEM; EDS	166 at 0.05°C (synthesized at 700°C)	50: ~ 0% at 0.1°C
LiFePO ₄ /C composite	Huang et al. [288]	Stearic acid-assisted rheological phase reaction at 600°C for 4 h	XRD; SEM; TEM; SAED	160 at 0.5°C 155 at 1°C	200: <3% at 2°C
Carbon-coated LiFePO ₄	Dong et al. [81]	Solid-state reaction at 650-800°C for 10 h followed by slow cooling	XRD; SEM	156.7 at 0.1°C (synthesized at 650°C)	50: 3.5%
Carbon-coated LiFePO ₄	Shin et al. [84]	Mechanochemical activation method	XRD; SEM; TEM; EIS	150 at 0.05°C 135 at 1°C	0.1–0.3 mAhg ⁻¹ /cycle
Carbon-coated nanocrystalline LiFePO ₄	Zhecheva et al. [86]	Freeze-drying method	XRD; BET; SEM; XPS	122 at 0.1°C and 500°C	85: 11.5%
SiO ₂ -coated LiFePO ₄	Li et al. [289]	Sol-gel method	XRD; TEM; SEM; EIS	158 at 0.1°C and 55°C 145 at 1°C and 55°C	100: 6% at 0.5°C and 55°C
ZrO ₂ -nanocoated LiFePO ₄	Liu et al. [290]	Precipitation method	XRD; TEM; SAED	146 at 0.1°C 97 at 1°C	100: 1.8% at 0.1°C 100: 3.1% at 1°C
CeO ₂ -coated LiFePO ₄ /C	Yao et al. [291]	Commercially LiFePO ₄ /C powder	XRD, SEM, TEM; EDS; EIS	153.8 at 0.1°C and 20°C 99.7 at 0.1°C and -20°C	30: 1.4% at -20°C
Ru-doped LiFePO ₄ /C	Wang et al. [292]	Rheological phase reaction at 350°C for 10 h and at 750°C for a few hours	XRD; SEM; EIS	156 at 0.1°C	30: ~ 0%
Cu-doped LiFe _{1-x} Cu _x PO ₄ /C (x=0, 0.01, 0.015, 0.02, or 0.025)	Chang et al. [293]	Solid-state reaction at 500°C for 6 h then at 750°C for 18 h	XRD; SEM	150 at 0.1°C (x=0.02) 127 at 2°C	-
Co-doped LiFe _{1-x} Co _x PO ₄	Zhao et al. [294]	Hydrothermal synthesis	XRD; XPS; TEM; EDS; Raman spectroscopy	170 at 0.1°C (x=1/4)	-
LiAl _{1-x} V _{1-x} PO ₄ F (x=0, 0.03, or 0.06)	Zhong et al. [295]	Carbothermal reduction	XRD; SEM	118 at x=0.03	30: 14.4%
Al-doped LiAl _{1-x} V _{1-x} PO ₄ F (x=0, 0.03, or 0.06)	Shengkui et al. [295]	Carbothermal reduction process	XRD; SEM	~118 (x=0.03)	30: ~ 14.4%
Li ₃ Fe _{2-2x} Ti _x Mn _x (PO ₄) ₃ /C (0 ≤ x ≤ 0.2)	Sun et al. [296]	Solid-state reaction at 850°C for 80 h followed by milling at 220 rpm for 10 h	XRD; SEM	112.3 (x=0.1)	20: 25.8%
LiFePO ₄ powders	Meligrana et al. [297]	Hydrothermal synthesis	XRD; ICP-AES, BET; SEM	~ 137 at 0.5°C ~ 130 at 1°C	20: ~ 0 at 0.5°C 20: ~ 0 at 1°C
LiFePO ₄ /C nanocomposites	Fey et al. [298]	Combination of carbothermal reduction and molten salt	XRD; SEM; TEM; DLS; EIS; SAED; EDS; Raman spectroscopy	141 at 0.2°C	-
LiCoPO ₄ /C	Xing et al. [85]	Sol-gel method	XRD; SEM; TEM; EIS	136.2	30: 32%
LiCoPO ₄ /C nanocomposites	Doan and Tangiuschi [299]	Combination of spray pyrolysis and wet ball-milling techniques followed by heat treatment	XRD; SEM; TEM; TG-TDA; EDS	109 at 0.05°C 142 at 20°C	40: 13% at 0.1°C
Vanadium-based compounds					
LiV ₃ O ₈	West et al. [121]	Freeze-drying and spray-drying	XRD; TGA	-	50: 0.5%/cycle

$\text{Li}_6\text{V}_{10}\text{O}_{28}$	Xie et al. [151]	Hydrothermal synthesis and annealing dehydration treatment	XRD; TGA; TG-TDA; ICP-AES; SEM; TEM	132	15: ~ 24%
LiV_3O_8	Xiong et al. [120]	Spray-drying method	XRD; SEM	340.2 at 25 mA g^{-1}	100: 15.2% at 125 mA g^{-1}
LiV_3O_8	Kannan and Manthiram [105]	Low-temperature solution dispersion method	XRD; SEM; DSC; TGA	~ 250 at 60°C	100: ~ -4%
$\text{Li}_3\text{V}_2(\text{PO}_4)_3/\text{C}$	Ge et al. [300]	Sol-gel method	XRD; SEM; TEM; EIS	179.8 at 700°C (d=30-50 nm and L= ~ 800 nm)	50: ~ 21%
$\text{Li}_3\text{V}_2(\text{PO}_4)_3/\text{C}$	Wang et al. [140]	Electrostatic spray deposition	XRD; SEM; TEM		
$\text{Li}_3\text{V}_2(\text{PO}_4)_3/\text{C}$	Yuan et al. [301]	Sol-gel method	XRD; SEM; TEM	127.9 at 1°C (synthesized at at 750°C) 124.1 at 5°C (synthesized at at 750°C)	100: ~ 0% at 1°C and 5°C
$\text{Li}_3\text{V}_2(\text{PO}_4)_3/\text{C}$	Chen et al. [302]	Carbothermal reduction process	XRD, FTIR, XPS, EIS	122 at 0.5°C at -25°C	100: 5% at 20°C
Cr-doped LiV_3O_8	Feng et al. [122]	Sol-gel method	XRD; SEM	269.9 at 150 mA g^{-1}	100: 5.6%
Polypyrrole- LiV_3O_8 composite	Feng et al. [123]	Solution dispersion in ethanol followed by co-heating process	SEM	292 at 40 mA g^{-1} (20% PPY)	40: ~ 14
Mixed amorphous-nanocrystalline LiV_3O_8	Shi et al. [127]	RF magnetron sputtering	XRD; TEM; SEM; SAED	382	100: 21.2%
$\text{LiV}_3\text{O}_8/\text{C}$ nanosheet	Idris et al. [129]	Hydrothermal synthesis followed by carbon coating	XRD; TGA; SEM; TEM; EIS	227 at 0.2°C	100: 14.5%
LiV_3O_8 nanocrystallites	Li et al. [124]	Solid-state reaction at 120°C for 20 h with intermittent grinding	XRD; SEM; TEM; TGA; TG-TDA	342 at 300°C	30: 11.7%
nanoporous LiV_3O_8	Ma et al. [126]	Tartaric acid-assisted sol-gel process	XRD; SEM; TEM	301 at 40 mA g^{-1}	50: 3.7%
Polypyrrole- LiV_3O_8 composite	Tian et al. [125]	Oxidative poly-merization of pyrrole using ferric chloride	XRD; FTIR; SEM	169 at 0.5°C	50: ~ 0%
LiV_3O_8 nanorods	Xu et al. [128]	Hydrothermal synthesis	XRD; TGA; TEM; FTIR	302 at 300°C	30: ~ 8%
$\text{Li}_3\text{V}_2(\text{PO}_4)_3$ nanocrystals-graphene oxide nanosheets	Rui et al. [138]	Modified Hummers method	XRD; TEM; SEM; TGA; Raman spectroscopy	128 at 0.5°C	100: ~ 0%
amorphous $\text{Fe}_2\text{V}_4\text{O}_{13}$	Si et al. [128]	Liquid precipitation method	XRD; SEM	235	40: ~ 14.5%
$\text{Mn}_x\text{V}_2\text{O}_5$ (x=0.02, 0.04, 0.09, or 0.19)	Park [303]	Sol-gel method with ion exchange resin	XRD; SEM; TGA	120 (x=0.09)	50: ~ 0%
Nanostructured composites					
K_2FeO_4	Wang et al. [304]	Hypochlorite oxidation method	XRD; CCD	~ 350	50: ~ 35%
LiFePO_4 powder	Ogihara et al. [250]	Spray pyrolysis	SEM; XRD; BET; AAS	150	500: 16%
LiF/Fe nanocomposites	Li et al. [305]	Mechanical ball-milling method	XRD	~ 568	20: 46%
Polypyrrole-sulfur nanocomposites	Liang et al. [306]	Self-degraded template for polypyrrole followed by co-heating process	XRD; FTIR; TEM; SEM; BET; TG-DSC	1151.7 (30 wt% sulfur)	80: 43.6% (30 wt% sulfur)
$\text{LiMn}_2\text{O}_4/\text{CNT}$ nanocomposites	Ding et al. [73]	<i>In-situ</i> hydrothermal method	XRD; SEM	116 at 1°C 84 at 10°C	100: 0.009% at 1°C 100: 5.8% at 10°C
$\text{LiMn}_2\text{O}_4/\text{CNT}$ nanocomposites	Xia et al. [307]	<i>In-situ</i> hydrothermal method	XRD; TEM; SEM; TGA	124 at 1°C 106 at 10°C	500: 8% at 1°C 1000: 23% at 10°C
LiFePO_4 /multiwalled CNT nanocomposites	Li et al. [308]	Solid-state reaction at 700°C for 12 h and hydrogen arc discharge method followed by mixing	XRD; TEM; SEM; EIS	155at 0.1°C 147 at 1°C	50: ~ 5% at 0.1°C 50: ~6% at 1°C
$\text{Li}[\text{Li}_{0.2}\text{Mn}_{0.54}\text{Ni}_{0.13}\text{Co}_{0.13}]\text{O}_2\text{-V}_2\text{O}_5$ composites	Gao et al. [110]	Co-precipitation method	XRD	300 at 0.05°C (10 wt% V_2O_5)	25: ~ 12%
V_2O_5 /polypyrrole composites	Ren et al. [104]	Sol-gel method using	XRD; FTIR; SEM; TGA	271.8 at 0.1°C (2.5 wt% PPY)	50: 17.1%

TEOS/[Bmim][NTf ₂]/Carbon-titania composites	Wang and Dai [165]	Sol-gel method using	STEM microscope; Micromeritics Tristar analyzer; BET; TEM, XRD, XPS	-	-
Advanced Cathode material					
PTMA	Nakahara et al. [171]	radically polymerizing	SEM,	-	1000: 11%
TCNQ/DDQ/THBQ	Hanyu and Honma [309]	Blending quinonic compounds and drying by vacuum	SEM;XRD	215.8 at room temperature	170: 27% at 0.2°C
Ellagic acid	Goriparti et al. [170]	Lithiation, delithiation; intercalation, deintercalation	FT-IR; NMR; LCMS, TGA, DSC, XRD, SEM	50	10: 25%
PAN/ sulfur	Wang et al. [172]	Heating process	TEM; XRD; FTIR; XPS	850 at	50: 5%/month
PEO	Marmorstein et al. [173]	Mixing powder	Electrochemical characterization	1600 at 90-100°C	10: 30%
Li ₂ S	Yang et al. [174]	Ball milled	SEM	800	10: 0.25%
Conversion Cathodes					
Metal Fluoride					
FeF ₃ /C, CMFNCs	Badway et al.	High energy mechanical milling	XRD, transmission electron microscopy	200 600 at 70°C	50: 10%
FeF ₃ /C	Li et al. [175]	Ball milling	XRD; TEM	712	10: 16%
(MF ₃) FeF ₃ /TiF ₃ and VF ₃	Arai et al. [178]	Lithiated; delithiated	XRD	140 at 25°C	35%
Li _{1-x} Ni _{1-x} O _{2-y} F _y	Kubo et al. [179]	Solid state reaction	XRD	240	30:-
Li ₄ Mn ₅ O _{12-n} Fn	Choi and Manthiram [180]	Solid-start reaction	XRD	150 at 500 and 600°C	50:-
LiAl _y Mn _{2-y} O _{4-z} F _z /LiAl _y Mn _{2-y} O ₄	Xiaomei et al. [181]	melt-impregnation method at 750°C	XRD	118	100: 25%
LiMn _{1.8} Li _{0.1} Ni _{0.1} O _{4-x} F _x	Matsumoto et al. [182]	Solid-state reaction	X-ray absorption near-edge structure (XANES)	Around 88 and 93	-
Li ₂ O/ VF ₃ and TiF ₃	Poizot et al. [257]	Li-alloying, solid-state	-	700 500-600	100: 0%
Carbon Fluoride					
FeF ₃ , FeF ₂ , and BiF ₃	Amatucci et al. [185]	solid state prelithiation process resulted	XRD, DMC, and TEM	243	0.18 at 24°C and
(CF) _n /(C ₂ F) _n	Watanabe [186]	Mild reaction conditions	XRD;ESCA spectra; DTA	-	-
IF ₅ / HF	Yazami [187]	RT and HT	XRD; IR spectroscGpy	-	-
Graphite	Wang et al. [188]	graphene paper prepared chemically with graphite	SEM	298 680 528	50: 19% 2: 87%
LiBF ₄	Yazami et al. [190]	Solid state	EPR; XRD; Raman spectroscopy; TEM	865	-
IF ₅ , NbF ₅ , TaF ₅ , MoF ₆ , WF ₆ , ReF ₆ , BF ₃	Hamwi et al. [191]	prepared by reaction of IF 5 with excess fluorine at 300°C	XRD; IR spectroscopy studies	-	-
CFx(RT)/LiC104	Hamwi et al. [192]	-	IR spectroscopy; XRD	667	60%
Fluorophosphates					
LiFePO ₄ /LiFe _{0.9} Mg _{0.1} PO ₄	1. Barker et al. [1]	Novel carbothermal reduction (CTR)	XRD	150	-
nano-carbon coated LiVPO ₄ F	2. Zheng et al. [198]	chemical lithiation; ball milling	XPS;XRD;TEM	143	50:-
LiVPO ₄ F	3. Li et al. [199]	sol-gel, calcinations	XRD;TEM	130	30: 4.6%
LiVPO ₄ F/C	4. Wang et al. [200]	Annealing	XPS; TG-DTA;XRD	136 at 0.1°C rate 123 at 1°C rate.	100: 4.7%
LiVPO ₄ F/ LiCoO ₂	5. Gover et al. [196]	carbothermal reduction	DSC	140	300 at 45°C 280 at 60°C
LiVPO ₄ F/C	6. Zhang et al [201]	Hydrothermal method	SEM; XRD; TEM	143	50: 2.8%
Na ₃ V ₂ (PO ₄) ₂ F ₃ / VPO ₄ and NaF	7. Gover et al. [202]	Solid state carbothermal reduction	XRD	120	220:-
NaVPO ₄ F	8. Zhao et al. [203]	Sol-gel method; heat treatment	XRD; SEM	117.3 and 106.8	100: 1.3%
Na ₃ V ₂ (PO ₄) ₂ F ₃	9. Jiang et al. [204]	Sol-gel method; heat treatment	TGA; TEM	127 and 117	30:-
Na ₃ V ₂ (PO ₄) ₂ F ₃	10. Song et al. [205]	carbothermal reduction at 700°C	SEM; XRD	188	37: 3.2%

$\text{Na}_{1.5}\text{VOPO}_4\text{F}_{0.5}$	11.	Sauvage et al. [206]	Tuning	XRD; FTIR; Rietveld analysis; atomic absorption measurements; stoichiometry; structure	-	50
$\text{Li}_{0.5}\text{Na}_{0.5}\text{MnFe}_2(\text{PO}_4)_3 / \text{Li}_{0.75}\text{Na}_{0.25}\text{MnFe}_2(\text{PO}_4)_3$	12.	Trad et al. [208]	Sol-gel method	XRD; neutron diffraction	96	22%
LiFePO_4F	13.	Huang et al.	Sol-gel; Ceramic	XRD; SEM	145	30: 4.7% and 3.9%
LiFePO_4F	14.	Ramesh et al. [218]	Solid-state	EDX; SEM; XRD; IR spectrum	145	40 at room temperature and 55°C
LiFePO_4F	15.	Wang et al. [219]	Novel sol-gel	EIS; galvanostatic	145	30: 5%
$\text{NaLiFePO}_4\text{F}$	16.	Khasanova et al. [213]	Annealing	XRD; EDX	113	6:-
$\text{Li}_2\text{CoPO}_4\text{F}$	17.	Truong et al. [215]	Two-step method combining a sol-gel route; solid state reaction	XRD; TEM; Raman; FTIR spectra	91	20: 25.3%
$\text{Li}_2\text{Co}_4-x\text{Fe}_x\text{PO}_4\text{F} / \text{Li}_2\text{Co}_4-x\text{Mn}_x\text{PO}_4\text{F}$	18.	Khasanova et al. [216]	Freeze-drying or ceramic techniques; ball-milled	XRD; SEM	75	10: 83%
Fluorosulfates						
LiFeSO_4F	19.	Ati et al. [221]	Ceramic preparation; ball-milled	XRD	85	25: 5%
$\text{LiFeSO}_4\text{F}_2$	20.	Reynaud et al. [223]	Electronically conductive coatings, high-energy ball milling	XRD, EDX	88	-

XRD: X-Ray Diffraction, BET: Brunauer-Emmett-Teller, TGA: Thermogravimetric Analysis, SEM: Scanning Electron Microscopy, EDS: Energy Dispersive X-Ray Spectroscopy, TEM: Transmission Electron Microscopy, DSC: Differential Scanning Calorimetric Analysis, FTIR: Fourier Transform Infrared Spectroscopy, SAED: Selected Area Electron Diffraction, HT: High Temperature, EIS: Electrochemical Impedance Spectroscopic, TOF-SIMS: Time-Of-Flight-Secondary Ion Mass Spectroscopy, CCD: Charge-Coupled Device, ICP-AES: Inductively-Coupled Plasma Atomic Emission Spectroscopy, EPMA: Electron-Probe X-Ray Microanalyzer, TG-TDA: Thermogravimetric/Differential Thermal Analysis, AAS: Atomic Absorption Spectroscopy, TG-DSC: Thermogravimetric/Differential Scanning Calorimetry, DLS: Dynamic Light Scattering

Table 1: Comparison of synthesis method, characterizations, specific capacity and cyclability of different cathode materials.

the safe limits, costly to manufacture and have aging issues, especially in hot places.

Lithium-ion rechargeable batteries have great achievement because of their characteristics as high energy density, long-term stability and its effectiveness as a solution for huge applications. This journal is completed to motivate reviewers who are interested in LIB, types of cathodes specifically and looking for environment-friendly, inexpensive and charge/discharge long-term cycles materials. Also, open the challenges front of researchers to discover new materials with better properties, characteristics, and features for LIB.

Electrochemistry

The basic working mechanism based on which LIBs functions is associated with the transfer of lithium ions from the positive electrode (cathode) to the negative one (anode) and vice versa. During discharging process, lithium ions travel through an electrolyte, often an organic solution of lithium salt such as LiPF_6 , from the cathode side to the anode side. The exact opposite occurs during charging as an external current is applied. Although all LIBs work according to this basic mechanism, there are two different processes by which lithium ions associate with cathodes or anodes: intercalation and reversible chemical reaction or alloying. Since the focus here is on the cathode side, a straightforward explanation of both processes follows.

Lithium transition metal oxides in the form of LiMO_2 or LiM_2O_4 , where M is usually Co, Ni or Mn, allow only for the intercalation of lithium ions around potential of 4 V [4]. Reversible intercalation of lithium ions virtually without structural changes of cathode materials makes the operational safety of LIBs superior to other types of secondary batteries such as lead acid and metal hydride nickel batteries. From other hand, in reversible alloying, insertion and extraction of lithium into the crystal structure of cathode with changes in the volume of the structural.

Reversible conversion reaction are lithiation reactions, it's proceed rapidly where the structural's are well-dispersed by lithium to metal structure (Figure 2) [5].

Cathode materials

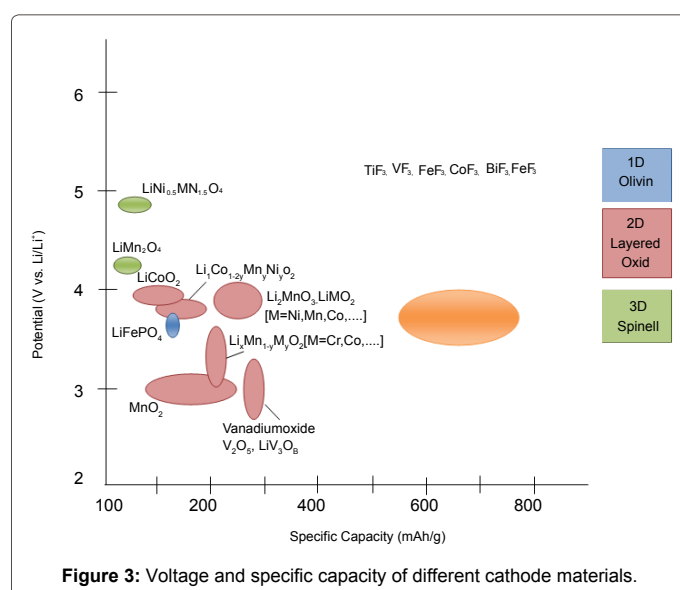
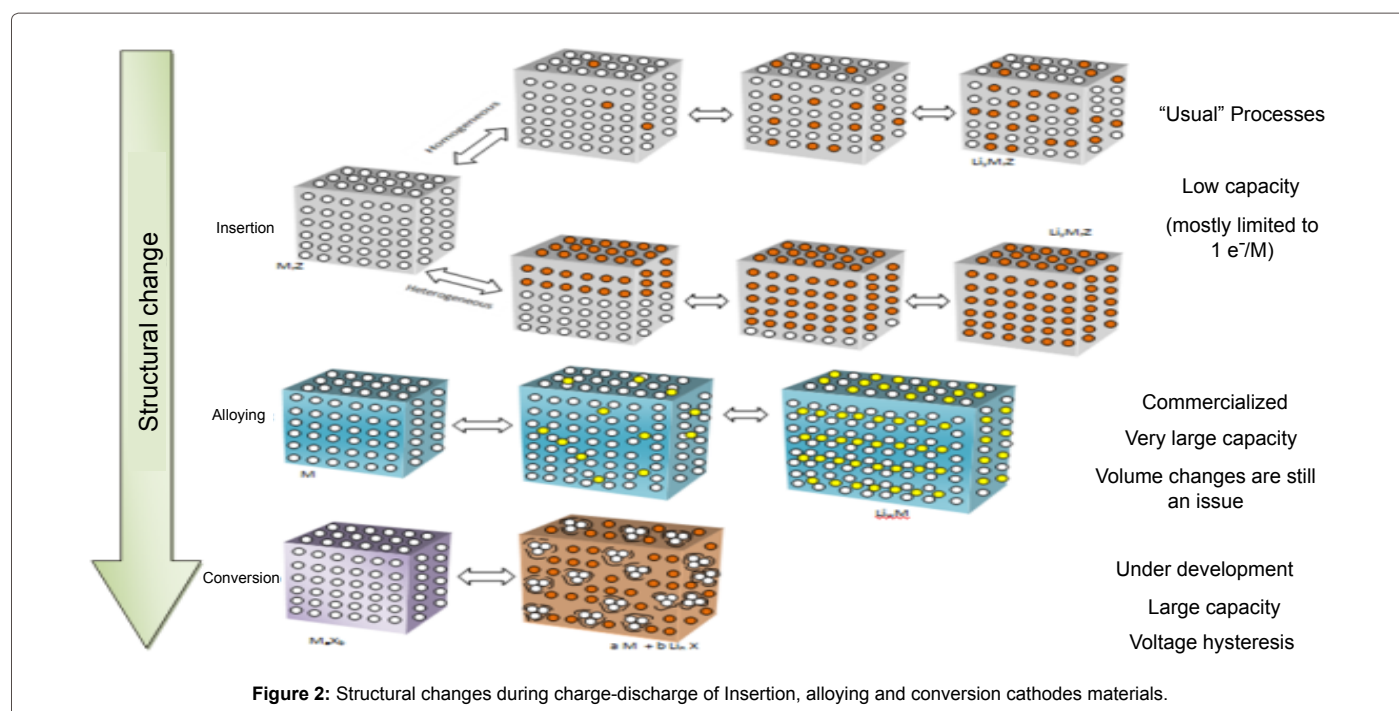
Despite the fact that layered LiCoO_2 has been the dominant cathode materials in commercialized LIBs, many important alternatives have attracted many researchers for potential use. Such substitutes, some of which have been already introduced to the market, include layered LiNiO_2 and LiMnO_2 along with their derivatives such as $\text{LiNi}_x\text{Co}_y\text{O}_2$, $\text{LiCo}_x\text{Ni}_y\text{Mn}_{1-x-y}\text{O}_2$, spinel-structured LiMn_2O_4 along with its derivatives such as $\text{LiNi}_x\text{Mn}_{2-x}\text{O}_4$ and $\text{LiCr}_x\text{Mn}_{2-x}\text{O}_4$, and olivine-structured LiFePO_4 . However, for any of these substitutes to be widely adapted, some challenges have to be overcome (Figure 3).

Layered Lithium Transition Metal Oxides

The layered lithium transition metal oxide with the formula LiMO_2 , where M=Co, Mn, Ni or a combination of two or more, have been arguably the most successful category of cathode materials for LIBs. Their superior electrochemical behavior can be ascribed to their layered structure which allows for a large number of diffusion paths for lithium ions (Figure 4).

LiCoO_2

As stated earlier, LiCoO_2 is the earliest and the most commonly-used cathode material for commercial LIBs. Suggested first by Mizushima et al. [6] in 1980, this material has several desirable features including high discharge potential, low molecular weight, high energy capacity, good charge/discharge performance, relative ease of synthesis and treatment, and stable and high discharge voltage [7]. However, extensive research has been conducted during the last two decades to find cathode materials with larger capacity and higher potential than LiCoO_2 . This was further motivated by the high cost, chemical hazards, and the environmental impact associated with cobalt. The preparation of LiCoO_2 was done by means of solid state reaction [8-10], sol-gel technique [11,12], ultrasonic spray pyrolysis process [13], combustion synthesis [14,15], co-precipitation method [16], molten salt synthesis



[17], freeze-drying method [18], complex formation method [19], hydrothermal synthesis [20], mechanochemical, and microwave synthesis [21,22], and other methods. Depending on the synthesis method, LiCoO_2 could have either hexagonal layered for high-temperature LiCoO_2 or cubic spinel-like structure for low-temperature LiCoO_2 [23]. A comprehensive review of high- and low-temperature synthetic methods and their effects on the electrochemical properties of LiCoO_2 was conducted by Antolini [23].

Despite its high theoretical capacity of 274 mAhg^{-1} , reported practical discharge capacities of LiCoO_2 are relatively low, in the range of $135\text{--}150 \text{ mAhg}^{-1}$ [14,18,24], only 50-55% of its theoretical capacity. In order to enhance the ionic conductivity and cycling performance of the cathode, some approaches such as carbon coating [11,25], coatings with oxide

compounds such as Al_2O_3 , ZnO , and LiTiO_{12} , and cationic doping on aluminum [26], chromium [27] and silver [28] have been applied. Among these, Al doping has received the greatest deal of attention as it has been proven to produce significant improvement on the capacity retention of upon cycling, which was attributed to an increase of the diffusion coefficient of lithium ions in Al-doped cathodes [26].

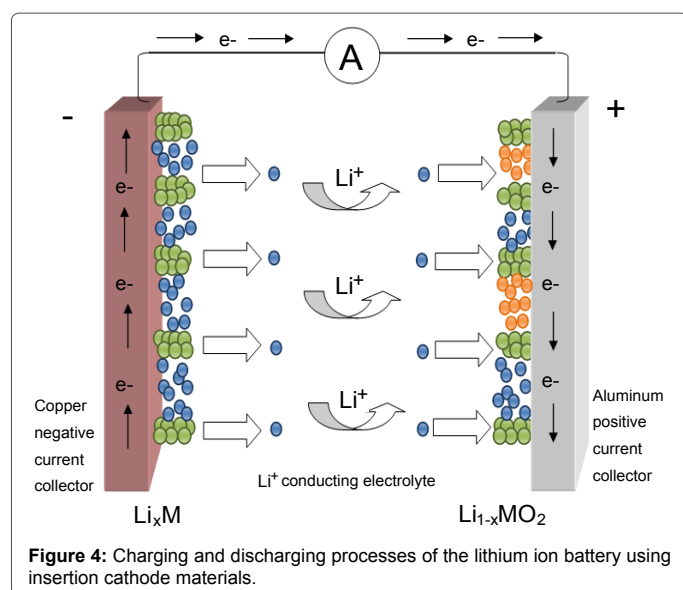
Various oxide coatings of LiCoO_2 cathodes have been studied including AlPO_4 , Al_2O_3 [29,30], CePO_4 , FePO_4 [16], Li_2CO_3 , $\text{Li}_2\text{PO}_2\text{N}$ [31], $\text{Li}_4\text{Ti}_5\text{O}_{12}$ [32], MgO [33], SrHPO_4 , SrO [34], SnO_2 , TiO_2 [34,35], ZnO [36], and ZrO_2 [35,37]. Having achieved a mixed record of success, some coatings remarkably increased the initial discharge capacity of LiCoO_2 as high as 190 mAhg^{-1} while others significantly enhanced the cycling behavior. For instance, Lu et al. [11] reported an initial discharge capacity of 195 mAhg^{-1} for Al_2O_3 -coated LiCoO_2 (1.0 wt% Al_2O_3) prepared by *in situ* sol-gel method with a good capacity retention upon cycling (85% after 30 cycles) [12]. In another study by Li et al. [16], 11.3% capacity loss was reported after 400 cycles for 3.0 wt% FePO_4 -coated LiCoO_2 cathodes. Other coatings which have produced significant improvements on the electrochemical behavior of LiCoO_2 include ZnO [38,39], ZrO_2 [35], $\text{Li}_2\text{PO}_2\text{N}$ [31], and $\text{Li}_4\text{Ti}_5\text{O}_{12}$ [32].

LiMnO₂

Although LiMnO_2 has been proposed as a cathode material in LIBs almost as early as LiCoO_2 , its use has not spread mainly due to performance limitations such as low capacity, difficulty of mass production, and power charge/discharge performance, especially at high temperatures. However, years of extensive research has led to significant improvement of its performance. Compared to LiCoO_2 , LiMnO_2 has major advantages such as high safety and low cost which make it a promising substitute in the future.

LiNiO₂

One of the early cathode materials to be explored was lithium nickel oxide (LiNiO_2) which has a comparable layered structure and



charge-discharge characteristics to those of LiCoO_2 . Although nickel-based cathodes are currently feasible for commercial use, their major drawback is poor solubility in organic electrolyte solutions, particularly at high temperature. Also, synthesis and treatment of LiNiO_2 often require harsh temperature conditions which further limit its current use in commercial LIBs despite its superior capacity [40]. Nickel has higher energy density than cobalt does; 50% of lithium ions can be transferred between anode and cathode for cobalt at the maximum voltage of a typical battery (4.7 V), while 70% of lithium ions can be mobilized for nickel at only 4.2 V.

Derivative compounds

The electrochemical behaviors of various layered derivative compounds have been extensively studied by numerous research groups. For more than two decades, researchers have been working on developing derivatives of nickel, cobalt, and/or manganese oxides in order to enhance the stability and improve the electrochemical behavior of layered cathode materials. Some of these derivatives are as $\text{LiNi}_x\text{Co}_{1-x}\text{O}_2$, $\text{LiCo}_x\text{M}_{1-x}\text{O}_2$, $\text{LiNi}_x\text{M}_y\text{Co}_{1-x-y}\text{O}_2$ (where $\text{M}=\text{Al}$ or Mn). Substantial improvement on the cycling performance has been reported by optimizing the composition of these derivative materials. Also, since manganese is less expensive and safer to use than cobalt or nickel, these derivatives could provide low-cost alternatives.

Surface modification by either coating with metal oxides or doping with metal cations proved to be an effective method for improving the chemical stability of layered derivative compounds. For instance, Subramanian and Fey (2002) reported a significant enhancement of the cycling behavior by cationic doping of $\text{LiNi}_{0.7}\text{Co}_{0.2}\text{Ti}_{0.05}\text{Mg}_{0.05}\text{O}_2$ (~ 0% after 20 cycles) [41].

Spinel Lithium Transition Metal Oxides (LiMn_2O_4)

LiMn_2O_4

The spinel lithium manganese oxide LiMn_2O_4 has been one of the most prospective cathode materials as a non-toxic, environmentally-friendly, the high natural abundance of Mn and low-cost candidate [42]. This material has a theoretical capacity of 148 mAh/g for an equivalent weight (M) is 180.8 g/mol [43].

A more detailed study of the electrical, thermal, and structural properties of LiMn_2O_4 as well as the conduction mechanism for this material can be found somewhere else [44]. Many researchers [45-47], who investigated the electrochemical properties of LiMn_2O_4 reported discharge capacity in the range of 100-120 mAhg^{-1} which represent 67-81% of its theoretical capacity (148 mAhg^{-1}) [48]. A wide variety of synthetic approaches have been applied to develop spinel LiMn_2O_4 including solid-state reaction [49], sol-gel method [50], hydrothermal synthesis [42], combustion synthesis, solution-phase, flame-assisted spray technology [51], and templating method.

Within the spinel lithium manganese oxide system, two approaches have been often proposed to improve the structural stability and electrochemical performance of the system: cationic substitution and surface modification. Manganese-substituted spinels of the structural formula $\text{LiM}_x\text{Mn}_{2-x}\text{O}_4$ ($\text{M}=\text{Al}$, B, Cr, Co, Cu, Fe, Ga, Ge, Na, Ni, Ti, Sc or Zn) have been extensively investigated by different research groups to improve the cycling performance of LiMn_2O_4 [52-61]. Among these derivatives, $\text{LiNi}_x\text{Mn}_{2-x}\text{O}_4$ showed significant improvement on cycling behavior of LiMn_2O_4 . For example, Wang and Xiao et al. showed a relatively good cycling behavior and higher reversible capacity of $\text{LiNi}_{0.05}\text{Mn}_{1.95}\text{O}_4$ electrode prepared by sol-gel method [62]. It was reported that lithium substituted used to make almost all commercial manganese oxide spinel materials (LMOs), since the cycling behavior is improved than LiMn_2O_4 . The theoretical capacities of 100-120 mAh/g , slightly lower than that for LiMn_2O_4 [63].

Surface modification has been a second way to improve the electrochemical performance of LiMn_2O_4 . Different salts have been studied such as Al_2O_3 , AlPO_4 , Cr_2O_3 [64], La_2O_3 [65,66], Li_3PO_4 [67], SrF_2 [68], TiO_2 [69], ZnO [70,71] and ZrO_2 [69]. Further nanostructural modifications have been examined such as encapsulation of LiMn_2O_4 nanowires in ZnO nanotubes by Liu et al. [72] and homogenous dispersion of LiMn_2O_4 nanoparticles in carbon nanotubes (CNT) composites by Ding et al. [73]. The latter approach demonstrated a significant improvement on the cycling behavior of LiMn_2O_4 approaching 100% after 100 cycles [73].

Derivative Compounds

Much research work has been conducted on the electrochemical properties of derivatives of LiMn_2O_4 in order to enhance the specific capacity and power and optimize the operational range of temperature for the original spinel compound. Two derivatives which have attracted many researchers since the early 90's, $\text{LiNi}_x\text{Mn}_{2-x}\text{O}_4$ [74], and $\text{LiCr}_x\text{Mn}_{2-x}\text{O}_4$ [52,75,76], have demonstrated a remarkable improvement on the cyclability of spinel magnesium oxides.

Olivine Lithium Transition Metal Phosphates and Silicates (LiMPO_4 and LiMSiO_4)

LiMPO_4

The electrochemical behavior of olivine-structured transition poly anion compounds of the structural formulas of LiMPO_4 and LiMSiO_4 ($\text{M}=\text{Co}$, Fe, Mn, Ni or V) have been attracted a great deal of interest as potential cathode materials. A comparative study of lithium intercalation potential in olivine-structured transition metal compounds has been reported by Zhou et al. [77].

Olivine-structured compounds have several advantages over other cathode materials including its structure of material hardly changes while Li ion intercalation and deintercalation; (2). It holds a long voltage platform.

Among phosphate compounds, LiFePO_4 has received the greatest amount of attention due to a number of desirable features such as low cost, non-toxicity, and good thermal and chemical stability [78]. For example, the main drawback of lithium iron phosphates, LiFePO_4 , is their low electrochemical performance at room temperature due to low lithium ion diffusion and poor electronic conductivity [78]. In order to overcome this drawback, several material processing techniques, including solid solution doping in metals and nanocoatings of phosphate particles with carbon, have been proposed.

LiFePO_4 has been prepared through different synthetic methods as reviewed by Zhang et al. [79] such as solid-state reaction, sol-gel synthesis, hydrothermal synthesis, carbothermal reduction, microwave synthesis, and spray pyrolysis. A comparative study of the synthetic routes of LiFePO_4 and their effect of the electrochemical properties was conducted by Franger et al. [80].

Another widely-investigated phosphate compound that has recently attained strong interest is LiCoPO_4 . LiCoPO_4 has been prepared by means of solid-state reaction, sol-gel synthesis, co-precipitation, hydrothermal synthesis, optical floating zone method, radio frequency magnetron sputtering, spray pyrolysis, and microwave synthesis. Zhao et al. [81] reported an initial capacity of 156.7 mAhg^{-1} and a capacity loss of 3.5% after 50 cycles for LiFePO_4 prepared by solid state reaction at 650°C .

Yang et al. studied mesoporous FePO_4 as a potential cathode material reporting an initial discharge capacity of 160 mAhg^{-1} with a capacity loss of 10% after 20 cycles. A significant amount of research has suggested that the migration of lithium ions, and consequently charge/discharge performance LiFePO_4 , can be greatly enhanced by carbon coating [81,82-86].

LiMSiO_4

Another group of poly-anionic compounds, silicates, have been examined for their interesting electrochemical potential as cathode materials including $\text{Li}_2\text{MnSiO}_4$ [87-89], $\text{Li}_2\text{CoSiO}_4$ [90-92], $\text{Li}_2\text{FeSiO}_4$ [78,88,93-102], and LiFeSO_4F [103]. However, several limitations have to be overcome before a wide use of such materials becomes feasible.

Nanostructured Metal Oxides

Vanadium oxides

The vanadium-based oxides have attracted strong interest from researchers due to their good electronic conductivity, excellent chemical stability in polymeric electrolytes, and high energy density [104]. Since vanadium can exist in a number of oxidation states from 2^+ in VO to 5^+ in V_2O_5 , vanadium oxides could offer a wide range of capacities as cathode materials [105]. Vanadium oxides which have been studied as potential cathode materials in LIBs are VO_2 [106], V_2O_5 [104,106-117], V_3O_6 [118], $\text{V}_3\text{O}_7 \cdot \text{H}_2\text{O}$ [119-125], LiV_3O_8 [104,126-139], $\text{Li}_3\text{V}_2(\text{PO}_4)_3$ [140-146], V_4O_{11} [147], $\text{Fe}_2\text{V}_4\text{O}_{13}$ [148], V_6O_{13} [149,150], and $\text{Li}_6\text{V}_{10}\text{O}_{28}$ [151]. Chernova et al. [152] reviewed in detail the structural and electrochemical features of different vanadium oxides as well as the process of lithium insertion in them [153-156]. Among these oxides, V_2O_5 , LiV_3O_8 and $\text{Li}_3\text{V}_2(\text{PO}_4)_3$ have shown the most promising electrochemical behavior as indicated by their high discharge capacities and good capacity retention. $\text{Li}_3\text{V}_2(\text{PO}_4)_3$ has been previously reviewed along with other phosphate compounds.

V_2O_5

As layered compounds with a high theoretical capacity of 442 mAhg^{-1} , V_2O_5 compounds are among the most promising high-capacity

cathode materials under development. Several synthetic methods have been employed to prepare V_2O_5 including sol-gel method, solvothermal route, precipitation process, and electrodeposition. A novel synthetic approach by Pomerantseva et al. enabled for nanostructured V_2O_5 thin films by biotemplated synthesis using Tobacco mosaic virus particles [114].

Studies on V_2O_5 have shown high discharge capacity mostly in the range of $250\text{-}300 \text{ mAhg}^{-1}$ for V_2O_5 /polypyrrole composites which represent 57-67% of its theoretical capacity with good capacity retention (15-20% after 50 cycles) [104]. When the structure and composition of lithiated V_2O_5 nanocomposites are optimized, their electrochemical behavior can be significantly enhanced. For example, in a study by Semenenko et al. [153] thin $\text{Li}_x\text{V}_2\text{O}_5$ ($x \sim 0.8$) nanorods with thickness of $5\text{-}10 \mu\text{m}$ were synthesized using hydrothermal treatment of V_2O_5 gel and lithium ions [157]. An initial discharge capacity of 490 mAhg^{-1} was reported with capacity loss of about 18% after 50 cycles [158].

Despite these advantages of V_2O_5 compounds, two drawbacks still persist: low power density due to their intrinsic low ionic conductivity, and poor cyclability as a result of microstructural failure upon cyclic lithium ion intercalation-deintercalation [104]. Since thermal stability in polymeric electrolytes is one of the main advantages for vanadium oxide, a variety of conductive polymeric materials have been suggested to be used as hybrid hosts of V_2O_5 such as polypyrrole [104,112,114], poly(ethylene glycol) [116], polythiophene [109], polyphosphazene [159], and polyaniline [115]. Such polymeric hybrid materials have been found to increase the electronic conductivity of the original oxides and improve the cycling behavior by enhancing the microstructural stability. Among these polymeric materials, polypyrrole has been the most extensively studied due to its high electric conductivity once doped with oxidizing agents and good electrochemical activity with a theoretical capacity of 72 mAhg^{-1} [160].

LiV_3O_8

Another lithium vanadium oxide that has been widely investigated over the last two decades for its good electrochemical properties is LiV_3O_8 . Such good electrochemical properties include high discharge capacity, high specific energy density, and long cycle life [120].

A wide range of synthetic routes have been used to prepare LiV_3O_8 in different forms including spherical particles [120], polymeric composites [123,125], and nanostructured materials of various morphologies such as nanorods [128], thin films [127,161] nanocrystals [162], and porous nanoparticles [126]. The synthetic routes studied in literature include solid-state reaction [124], spray pyrolysis method [163], sol-gel process [126], hydrothermal synthesis [105,122], radio frequency magnetron sputtering [156,127], hydrothermal treatment [128], rheological phase reaction method [164] and microwave synthesis [165]. In order to enhance the electrochemical properties of the material synthesized by increasing surface porosity and lowering crystallinity, several techniques have been suggested such as ultrasonic treatment [166] and partial crystalline modification by introducing small amounts of H_2O , CO_2 and NH_3 [167]. Such techniques, although have improved the electrochemical behavior of LiV_3O_8 to some degree, need further optimization in order to produce satisfactory power and cycling efficiency and to be applicable in large-scale production systems [159].

It was found by several researchers that the synthetic routes affect greatly the capacity of LiV_3O_8 cathodes [159]. Even though the initial discharge capacity could even reach more than 100% of its theoretical capacity (280 mAhg^{-1}) [157] when certain nanostructuring techniques or polymeric alloying are applied. Idris et al. [129] reported an initial

discharge capacity of 227 mAhg⁻¹ with low capacity loss (~ 15% after 100 cycles) for LiV₃O₈/carbon nanocomposites prepared by hydrothermal synthesis followed by a carbon-coating process. Other researchers such as Feng et al. [123] and Tian et al. [125] obtained higher initial discharge capacity (~ 300 mAhg⁻¹) with good cycling efficiency (8-14% capacity loss after 30-40 cycles) for polypyrrole-LiV₃O₈ composites.

Coatings of Cathode Materials

A comprehensive review of surface coatings of cathodes in LIBs was conducted by Li et al. [163]. Such coatings were found to enhance the structural stability of the cathode material by limiting the contact with electrolyte solution, suppress phase transition, and stabilize the cations in their crystal sites. Nanocoatings of LiMn₂O₄ include SiO₂ [168-172].

Nanostructured Composites

Nanostructured carbon-oxide composites

Wang and Dai [165] developed an approach towards functionalized porous carbon-oxide composite materials by using ionic liquid (ILs) as solvents in nonhydrolytic sol-gel processing and this to limited and oxide-catalyzed carbonization of ILs trapped within an oxide framework. BET, TEM, XRD and XPS characterization measurements were applied.

Nanostructured polymer-oxide composites

Zhang et al. prepared LiFePO₄/C composite fibers by a combination of electrospinning and sol-gel techniques using Polyacrylonitrile (PAN) as an electrospinning media. XRD and SEM measurements were carried out to characterize the structure of the fibers formed [173].

Lithium/Air

Recently, a growing attention has been directed to lithium/air batteries which utilize mesoporous carbon as cathode materials. These devices represent a special category of LIBs as they enjoy very high energy density compared to other conventional types of lithium-ion batteries.

Organosulfur Materials

Nowadays, organosulfur compounds usually with the organothiol (-SH)/disulfide (S-S) redox couple, it's characterized by their ability to store big amounts of charges per unit mass to use as cathode materials for rechargeable lithium batteries and featuring of being safety, cheap, synthetic it easily and chemically stable. The theoretical energy of organosulfur compounds goes beyond that of as intercalation compounds, conducting polymers and conventional battery materials [166,167]. Organosulfur compounds containing disulfide bonds showed a high discharge capacity equal to 500 mAh/g [168].

Advanced Cathode Material

Organic cathodes

Most of the recent lithium batteries made of inorganic compounds as a cathode are produced from nonrenewable resources and because of that they are highly cost [174-177]. Scientists searched for another candidate to improve the power and energy density, safety of Li-ion cells and greener Li-ion batteries. Organic electrodes have been proposed as one of the best electrodes for Li-ion batteries due to its inherently flexible, non-toxic, cheap, abundant nature and also their limitation of cycle life, thermal stability, low energy density values

and rate capability led to a huge improvement of it [170]. Nakahara et al. proposed a high-energy organic cathode material; poly(2,2,6,6-tetramethyl-1-piperidinyloxy-4-yl methacrylate) (PTMA) for use in lithium rechargeable batteries, it is obtained good power capability, cycling efficiency (retaining more than half of original capacity after 1000 cycles), fast charging and discharging (less than 1.5 min) and can transfer specific capacity over 100 mAh.g⁻¹ [171].

Sulfur compounds Li₂S

Except the air cathode, the sulfur element has the cheapest cathode material for lithium batteries and top theoretical capacity density of 1672 mA/g between all known cathode materials [172]. Li₂S cathode material shows a great potential of high-performance rechargeable lithium batteries comparing with other resources of elemental sulfur in nature, like micro-batteries for power sources for electric vehicles and small-size electronic devices emphasizing high charge density [169]. However, because sulfur element has the dissolution of its reaction product polysulfides into the electrolytes and highly insulating nature, it cannot be used directly at low temperature as an electrode material for lithium batteries, which caused various problems, such as rapid fall of the capacity and short utilization of active material [178]. Yang et al. [174] found that Li₂S can be an active material and reached a large potential barrier (~ 1 V) at the beginning of charging by applying a higher voltage cutoff. It's obtained a greater initial discharge capacity (~ 800 mAh/g) which becomes stabilized after (10 cycles with around 500-550 mAh/g, ~ 0.25% per cycle capacity decay rate).

Conversion cathodes

To improve cathode materials, electrochemical conversion reactions have been used as another way to accomplishing the utilization of all the oxidation phases of a transition metal [179-182]. Fluorides metal are one of greatest transition metals that commonly studied in the research because of its stability and its ion considered as a strong and suitable ionic character of the (M-F bond) to transfer charges between two electrodes and produce high operating voltages and reversible capacity [176]. Iron fluoride (FeF₃) is one of the derivative of transition metal fluorides that safety and inexpensive, it is characterized by high theoretical capacity (712 mAh/g) [177,175]. In 1990, Arai et al. were the first researcher who reported the electrochemical activity of trifluorides with its high theoretical voltage and a specific capacity equal to 80 mAh/g [178].

Among various studies of fluorinated, fluorinated solvents are the most studied since it's used for increasing the safety and stability in LIB [183-186]. Different fluorine-doped intercalation cathodes are produced from fluorine which has been used as a dopant like layered transition metal oxyfluorides (Li_{1-x}Ni_{1-x}O_{2-y}F_y), spinel lithium manganese oxyfluorides (Li_{1+x}Mn_{2-x}O_{4-y}F_y) and orthorhombic lithium manganese oxyfluorides (Li_{1.07}Mn_{0.93}O_{1.92}F_{0.08}) [187].

Layered transition metal oxyfluoride: As have been mentioned before about layered lithium transition metal oxides owing to their electrochemical performance for secondary Li-ion batteries as high potential electrode materials. However, fluoride cathodes have the highest average voltage between conversion reactions [179].

Li_{1-x}Ni_{1-x}O_{2-y}F_y: Kubo et al. [180] discussed metal oxyfluoride as cathode materials by studying the synthesized of Li_{1-x}Ni_{1-x}O_{2-y}F_y, through solid state reaction that led to substitute nickel and oxygen positions for LiNiO₂. X-ray diffractometry and electrochemical technology were used to examine the cycle properties and show initial charging capacity around 240 mAh/g. Li_{1.08}Ni_{0.92}O_{1.9}F_{0.1} possessed

excellent features on charge/discharge cycling compared to LiNiO_2 and showed enhanced in the capacity retention during cycling [188-190].

Spinel lithium manganese oxyfluorides: Spinel lithium manganese oxide LiMn_2O_4 has been studied widely as cathode materials due to its features of being non-toxic, inexpensive and environmentally-friendly material. Spinel LiMn_2O_4 can deliver 120 mAh/g capacity. Choi and Manthiram [182] reported the synthesized of spinel $\text{Li}_4\text{Mn}_5\text{O}_{12-n}\text{F}_n$ oxyfluoride cathodes at 500 and 600°C by substituted F-ions from employing LiF for oxygen (O_{2-}) ions in the spinel $\text{Li}_4\text{Mn}_5\text{O}_{12}$. XRD, electrochemical cycling, and chemical analysis measurements were carried out to characterize $\text{Li}_4\text{Mn}_5\text{O}_{12-n}\text{F}_n$ and it has been found the useful of oxyfluoride cathodes to increase and enhance the capacity [181]. However, many different types of research discussed different ways to improve the Spinel lithium manganese oxyfluorides [182-184].

Metal fluorides: Conversion reactions start to attract attention recently, like fluorides, nitrides, sulfides and phosphides. Fluoride showed a great reversible cathode electrode that used to react with Li at 2.5 V and produced 800 mWh/g of energy density [191-193]. Poizot et al. [179] studied the mechanism of Li reactivity depend on involves the formation and decomposition of Li_2O , escorting the oxidation and reduction of metal nanoparticles. However, in the past and due to metal fluorides characteristic such as insulating nature and apparent irreversibility in structural conversion [175], it has been ignored as rechargeable cathode materials for lithium batteries. Malini et al. [186] reported two ways to exploit the electrochemical efficiency of metal fluorides: first one, by mixing metal fluorides with conducting carbon materials to improve electrical conductivity and the second one, mechanical ball milling of metal fluoride to reduce the particle size [194-197]. VF_3 and TiF_3 are also transition metal fluoride, they demonstrated their effectiveness with Li and generated as high as 500-600 mAh/g [175]. Amatucci et al. [187] studied the basic reactions of alkali nitrides with metal fluorides by prepared pre-lithiation agent, Li_3N , to (FeF_3 , FeF_2 , and BiF_3) metal fluorides. XRD, DMC, and TEM measurements were carried out to characterize the structure of the nanocomposite product and showed 243mAh/g initial charge capacity.

Carbon fluorides: Carbon fluorides consider as a great theoretically materials for high energy batteries because of its high theoretical potential, low equivalent weight and also most of them produce a very low self-discharge and extraordinary stability which pays high attention for carbon fluorides [177].

Two main groups are classified carbon fluorides: high temperature (HT) up to 300°C and room or low temperature (LT). HT fabricated graphite fluorides compounds consisting of two stages, $(\text{CF})_n$ and $(\text{C}_2\text{F})_n$ respectively while LT fabricated fluorine-graphite intercalation compounds of CF_x [198,199]. Various studies have been studied the lithium batteries contained graphite fluoride as cathode material [200-205]. Therefore, the specific energy densities of covalent graphite fluorides of the (C-F bond) reached 900 W h kg^{-1} [191]. Many researchers have been discussed graphite fluorides in their both categories: HT [193,194], and at LT [195-197]. Electrochemical performance of LT fluorinated graphite's studied by Delabarre et al [198] where the compounds prepared at room temperature under fluorine gas, XRD, FT-IR, NMR and EPR measurements were carried out to characterize the electrochemical properties.

Fluorophosphates

New studies of cathodes materials for LIBs have been conducting about poly-anionic materials containing fluorine as part of their compounds, known as fluorophosphate. LiMPO_4F and $\text{Li}_2\text{MPO}_4\text{F}$

($\text{M}=\text{Fe}, \text{Ni}, \text{Co}$) were the general formulas of fluorophosphate containing Li and crystallized materials that have been discovered as high-potential cathode materials [199], and thermal stability [71] for rechargeable lithium batteries. Three structured compounds have been widely studies, LiVPO_4F , NaVPO_4F , and LiFePO_4F [63].

LiVPO_4F : Lithium vanadium fluorophosphate it is a new class of cathode material that produced high voltage, long cycle life, excellent thermally stability, stable crystalline structure with good quality [201-204]. Carbothermal reduction method used mainly to prepare LiVPO_4F , also it is synthesized through one or two-step solid reactions [201], chemical lithiation [206-210], sol-gel method [206] and post-annealing [211-215].

Theoretical capacity (156 mAh/g) with potential equal to 4 V was reported by Saidi and Barker [150]. Gover et al. [210] prepared LiVPO_4 as a cathode electrode in LIB using carbothermal reduction method, DSC, and electrochemical measurements were performed. The discharge capacity is about 140 mAhg⁻¹ for the positive electrode with average discharge voltage around 4V [200]. Other research done by Zhang et al. [208] depended on prepared $\text{LiVPO}_4\text{F}/\text{C}$ nanosheets with homogeneous carbon coating by applying a hydrothermal approach and calcinations respectively. X-ray diffraction, SEM, TEM measurements and electrochemical tests have been performed and showed initial discharge capacity (143 mAh/g) with potential between (3.0-4.5 V) [216].

NaVPO_4F : Sodium vanadium fluorophosphate characterized as a safer, economical and higher work potential comparing to other materials [209,210]. This includes $\text{Na}_3\text{V}_2(\text{PO}_4)_2\text{F}_3$ with theoretical capacity equal to 128 mAh/g [170,211,212], NaVPO_4F [209] and $\text{Na}_{1.5}\text{VOPO}_4\text{F}_{0.5}$ [213]. Generally, NaVPO_4F materials synthesis required VPO_4 as the reaction intermediate phase, also it's successfully synthesized by three strategies: first, solid-state which require high temperature, long time-consuming process and complex operation procedure [209,212], second ion exchange or third, hydrothermal approach that considered as a complicated system to collect the results of compounds after the procedures finished [209]. $\text{Na}_3\text{V}_2(\text{PO}_4)_2\text{F}_3$ classified as the most important among fluorophosphate materials due to the high theoretical capacity 192 mAh/g and its flexibility to be used as a cathode in both Li-ion batteries and Na-ion batteries [214]. While $\text{Na}_{1.5}\text{VOPO}_4\text{F}_{0.5}$ delivered through 3.6 and 4 V vs. Na^+/Na theoretical capacity equal to 156 mAh/g [217-221].

LiFePO_4F : Another option about olivine-type, favorite-structured lithium-metal-fluorophosphate as cathode material achieved a good alternative for LIBs [216]. In lithium iron fluorophosphate, one Li^+ can be cycled charge/discharge with the theoretical capacity of 153 mAh/g [222-225]. LiFePO_4F was conducted by Ramesh et al. [218] as a new adequate material of fluorophosphate because of Li^+ feature that migrates easily without any barriers. LiFePO_4F prepared by solid-state routes and produced a reversible capacity approximately 145 mAh.g⁻¹, with stable electrochemical cycling (40 cycles at room temperature and 55°C) [218]. In another study by Wang et al. [219] LiFePO_4F prepared by a novel sol-gel process, EIS and galvanostatic were unambiguously performed and showed reversible capacity equal to 145 mAh.g⁻¹, with outstanding cyclic performance and fast transfer of Li^+ comparing to raw LiFePO_4 . Other various fluorophosphate materials have been studied including $\text{Li}_2\text{FePO}_4\text{F}$ [218,226-230], $\text{Li}_2\text{MPO}_4\text{F}$ ($\text{M}=\text{Co}, \text{Ni}$) [231-235], and $\text{Na}_3\text{V}_2(\text{PO}_4)_2\text{F}_3$ Hybrid Ion Cathode [204,210].

Fluorosulfates

New material of cathode materials reported first by Sebastian et al. was LiMgSO_4F , they presented the benefit of LiMSO_4F ($\text{M}=\text{Mn}$,

Fe, Co) for redox extraction/insertion of lithium involving MII/MIII oxidation states [236-240]. Then reported by the Sauvage [213], who prepared in the tavorite type structure LiFePO_4OH . Investing the large electronegativity of sulfur and fluorine of LiMSO_4F in produce high-voltage cathode materials such as LiFeSO_4F transfer around 140 mAh/g with a high redox potential of 3.6 V [241-244]. Different Li metal fluorosulfate compounds (M=Mg, Mn, Fe, Co, Ni, Zn, Cu) were studied and shown a great electrochemistry and crystal chemistry counting on the type of metal ions and on the synthesis methods [227].

LiFeSO_4F : Ionothermal was the first method to produce LiFeSO_4F , it can be reused repeatedly for synthesizing although it is very high cost ionic liquid media [245-250]. LiFeSO_4F has been prepared through various synthetic processes such as solid-state reaction [140], polymer-assisted [228], tradition ceramic preparation [229], electronically conductive coatings, high-energy ball milling [251-255], and solvothermal reaction [224]. The recent synthesizing methods found that tavorite-type LiFeSO_4F has low thermodynamic stability, unstable at temperatures above 350°C, because of that, producing single-phase of tavorite LiFeSO_4F It is difficult to occur [103,256-259]. LiFeSO_4F has a theoretical capacity of 148 mA h/g and produced an energy density (543 Wh/kg) as good as LiFePO_4 (581 Wh/kg) [260-263]. Rosati et al. [264] reported the reversible capacity of approximately 85 mA h.g⁻¹ of LiFeSO_4F which prepared in the triplite structure by the ceramic method at 300°C [265-269]. LiFeSO_4F has numerous advantages as cathode material such as it can be prepared from plentiful $\text{FeSO}_4 \cdot n\text{H}_2\text{O}$ precursor and synthesized at low temperature [270-276].

LiMSO_4F (M=Co, Ni, Mn, Zn): Fluorosulfate polyanionic LiMSO_4F electrode materials represent a wide family of a good combine of properties, especially, both electrochemical and safety issues. LiMSO_4F showed a redox potential of 4.25, 4.95, and 5.25 V, respectively [277-281]. $\text{Li}_2\text{CoP}_2\text{O}_7$ is considered as a 4.9 V while $\text{Li}_2\text{NiP}_2\text{O}_7$ is considered as a 5.4-V cathode [231,281-285]. In contrast with $\text{Fe}^{2+}/\text{Fe}^{3+}$ redox reaction, $\text{Ni}^{2+}/\text{Ni}^{3+}$, $\text{Mn}^{2+}/\text{Mn}^{3+}$ and $\text{Co}^{2+}/\text{Co}^{3+}$ redox reactions do not take place within the explored galvanostatic cycling potential window [286-291].

LiMSO_4F (M=Co, Ni, Mn, Zn) synthesized at low temperature similarly to LiFeSO_4F by using solid-state [291-296] and polymer-assisted methods [297-300]. Other various fluorosulfates materials have been studied including $\text{Li}(\text{Fe}_{1-x}\text{M}_x)\text{SO}_4\text{F}$ (M=Mn, Zn), NaMSO_4F (M=Fe, Co, Ni, Mn, Mg, Zn, Cu), $\text{NaMSO}_4\text{F} \cdot \text{H}_2\text{O}$ (M=Fe, Co, Ni, Zn) [301-306].

Conclusion

Rechargeable batteries are considered of crucial role nowadays and become mandatory for most important electronics devices that most of the people use in communication, transportation, and monitoring. LIBs need an improvement in their characteristics for future applications where high energy and power density with long-term stability are required [307-310]. This paper summarized the characteristics of different types of cathode materials for LIBs and compare between their electrochemical performance such as specific capacity, thermal stability, synthesis method, and characterization techniques. The best cathode materials for LIBs should have high capacity, inexpensive, environment-friendly and charge/discharge long-term cycles for large and practical applications. However, as much as material exist, as well as challenges present, challenges of cathode materials include nanostructuring, switching from insertion to alloying and conversion materials, and improving cyclability and life types. Therefore, better understanding the mechanisms involved in charge-discharge of

different cathode materials will certainly help scientists to overcome volume changes and hysteresis phenomena encountered with alloying and conversion materials. LIBs are expected to reach more commercial production in the future with better improvements in energy density and capacity.

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