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 performances 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 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).

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
| Material | Research group (year) | Synthesis method | Characterization technique | Initial capacity (mAh/g) | Number of cycles: Capacity loss |
|----------|----------------------|------------------|---------------------------|--------------------------|--------------------------------|
| Layered metal oxides | | | | | |
| LixCoO2 (0 ≤ x ≤ 1) | Mizushima et al. [6] | Electrochemical extraction from the Li2O2 rocksalt structure | XRD | - | - |
| HT-LiCoO2 | Shlyakhitin et al. [18] | Freeze-drying method | XRD; SEM; TEM; SAED | 135-138 | 10-10.14% |
| LiCoO2 | Gan et al. [14] | Carbon combustion synthesis | XRD; SEM | 148 | 10.3% |
| Carbon-coated LiCoO2 | Cao et al. [25] | Commercial LiCoO2 powder with milling for 24 h at 300 rpm | XRD; SEM; TEM; EIS | 130 at 0.1°C | - |
| Carbon-coated LiCoO2 thin film with PVDF-HFP gel electrolyte | Park et al. [11] | Sol-gel process and Screen printing | XRD; SEM | 110 | - |
| ZnO-coated LiCoO2 | Chang et al. [26] | Plasma-enhanced chemical vapour deposition | XRD; SEM; AAS; TGA; DSC; BET; EIS | ~ 178 at 1°C | 30: ~33% |
| Al2O3-coated LiCoO2 | Lu et al. [10] | In situ sol-gel process followed by calcination at 1123 K for 12 h | XRD; SEM; TEM; XPS; DSC | 195 (1.0 wt% Al2O3) | 30: 15% |
| Li2O2-coated LiCoO2 | Yi et al. [32] | Commercial LiCoO2 powder and coating with sol-gel process | XRD; SEM | 142.7 (3 wt% Li2O2) | 40: 2.7% |
| FePO4-coated LiCoO2 | Li et al. [16] | Co-precipitation method followed by a high-temperature treatment | XRD; SEM; TEM; EIS; AAS | 146 (3 wt% FePO4) | 400: 11.3% |
| High-density spherical LiCoO2 | Ying et al. [24] | Controlled crystallization method | XRD; SEM | 148.4 at 0.2°C | 40: 2.4% |
| ZnO-coated LiCoO2 | Fang et al. [39] | Commercial LiCoO2 powders coated with wet chemical method | XRD; SEM; ICP-AES | ~ 188 at 0.1°C (0.2 wt% ZnO) | 30: 10.4% |
| ZnO-coated LiCoO2 | Fang and Duh [38] | Wet chemical method with a calcination process | XRD; SEM | ~ 185 (calcined at 650°C) | 30: 15.5% |
| SrO/Li2O/La2O3/Ta2O5/TiO2-coated LiCoO2 | Wang et al. [34] | Commercial LiCoO2 powders coated with Sol-gel method | XRD; SEM | ~ 140 at 1°C (1.0 wt% SrO/Li2O/La2O3/Ta2O5/TiO2) | 900: ~25% |
| LiNiO2 | Yamada et al. [241] | Solid-state reaction at 500-900°C for 5 h | XRD | 197 at 1°C (at 700°C in O2) | 12: ~0% |
| LiNiO2 | Kalyani et al. [21] | Microwave synthesis with O2 heat pre-treatment | XRD; SEM | 156.8 at 9.5°C (~12.5 wt% binary conductive additives) | - |
| Co-coated LiNiO2 | Sheng-wen et al. [24] | Co-precipitation method | XRD; SEM; TEM; XPS | 180 | 20: 0.07%/cycle |
| Ga-doped LiNiO2 | Nishida et al. [243] | Sol-gel process | XRD | 190 | 100: 5% |
| LiNi1-xMgxO2 (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% |
| LiCo1-xNixO2 (0 ≤ x ≤ 0.2) | Gummow and Thackeray [245] | Sol-gel process | XRD | ~178 (x=0.5) | 65: ~16% |
| Li2PO2N-coated LiCoO2 | Choi et al. [31] | RF magnetron sputtering method | EDS; SEM; DSC; EIS | ~ 185 at 0.2°C | 50: 16% |
| LiCoO2/LiNi0.8Co0.17Al0.03O2 | Lin et al. [236] | Commercial products | - | 186 (for pure LiNi0.8Co0.17Al0.03O2) | 20: 5% |
| LiCoO2 thin film | Park et al. [237] | Screen-printing method using ethyl-cellulose-based paste | SEM | 133 | - |
| LiCoO2/polyacrylonitrile/binary conductive additives composite | Zhang et al. [238] | Commercial products | XRD; SEM; TEM | 156.8 at 9.5°C (~12.5 wt% binary conductive additives) | - |
| poly(3,4-dioxyethylenethiophene)/LiCoO2 composite | Her et al. [239] | Electrochemical deposition process | DSC; SEM; EDS | - | 50: ~0% |
| Ag-doped LiCoO2 | Huang et al. [26] | Commercial LiCoO2 powder with milling for 4h in alcohol | XRD; SEM | 172.3 at 1°C 133.1 at 3°C | 50: 22.29% at 1°C 50: 4.88% at 10°C |
| LiMnO2 (0.4 ≤ x ≤ 0.7) | Pistoia et al. [240] | Solid-state reaction | XRD; TGA, TDA | ~108 (x=0.55) | 65: ~16% |
| LiNiO2 | Yamada et al. [241] | Solid-state reaction at 500-900°C for 5 h | XRD | 197 at 1°C (at 700°C in O2) | 12: ~0% |
| LiNiO2 | Kalyani et al. [21] | Microwave synthesis with O2 heat pre-treatment | XRD; BET; SEM | 163 | <5% |
| Co-coated LiNiO2 | Sheng-wen et al. [24] | Co-precipitation method | XRD; SEM; TEM; XPS | 180 | 20: 0.07%/cycle |
| Ga-doped LiNiO2 | Nishida et al. [243] | Sol-gel process | XRD | 190 | 100: 5% |
| LiNi0.5Mn0.5O2 | Abdel-Ghany et al. [246] | Wet-chemical method | XRD; SEM | 166 | 30: 5% at 900°C |
| Compound                  | Source                          | Synthesis/Merchandizing Method | Characterization Method(s) | Synthesis temp (°C) | Properties |
|---------------------------|---------------------------------|-------------------------------|-----------------------------|---------------------|------------|
| LiNi0.9Co0.1O2            | Shi et al. [247]                | Rheological phase reaction method | XRD; SEM                   | ~199 (synthesized at 800°C) | 15: 27.1%  |
| LiNi0.9Co0.1O2 powder     | Sivaparakash et al. [248]       | Solid-state reaction at 700-800°C for 15-48 h with intermittent grinding | XRD                     | ~ 157                | 20:~11%    |
| LiMn0.1Ni0.9O2.5O2        | Wang et al. [249]               | Solid-state reaction at 800-900°C | XRD; SEM                   | 180 (x=0.2 and y=0.25) | 50: 15%    |
| LiCo0.3Ni0.7O2 powder     | Oghihara et al. [250]           | Spray pyrolysis                 | SEM; XRD; BET; AAS         | 169                  | 500: 20%    |
| LiNi0.9Mg0.1O2            | Subramanian and Fey [41]        | Solid-state reaction at 800°C for 12 h | XRD; SEM; ICP-AES          | 149                  | 30: ~2% (Mg-doped) |
| LiNi0.8Co0.2O2 powder     | Solid-state reaction at 700-800°C for 15-48 h with intermittent grinding | XRD                        | ~170 (x=0.1)              | 30: ~ 12%          |
| Li(Al,Co)2O4 (0 ≤ x ≤ 0.3)| Myung et al. [26]              | Emulsion-drying method          | XRD; TEM                   | 110 (x=0.1)         | -           |
| Li(Ni,Co)2O4 (0 ≤ x ≤ 0.3)| Huang et al. [252]             | Solid-state reaction at 200°C for 20 h and at 750°C for 24 h with intermittent grinding | XRD                     | 153                  | 15% at x=0.1 |
| LiNi0.9Mg0.1O2 (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%    |
| LiMn0.1Ni0.9O2            | Ren et al. [254]                | Solid-state reaction at 900°C for 20h | XRD; SEM                   | ~170 at x: 1/3     | 20~0        |
| LiAl0.05Ni0.95-xCoxO2     | Zhu et al. [253]                | Low temperature combustion followed by annealing treatment | XRD; SEM; BET; EIS       | 174 at 1°C          | 10: <5%     |
| LiAl0.05Ni0.95-xCoxO2     | Kim et al. [257]                | Co-precipitation method at 1000°C for 10 h | XRD; SEM; EIS             | 173.8                | 50: 10.9%   |
| LiAl0.05Ni0.95-xCoxO2     | 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 |
| LiMn2CoO4 crystalline     | Son and Cairns [259]           | Sol-gel method                  | XRD; TEM                   | 164.1 at 0.1°C    | 20: ~ 0% at 0.1°C  |
| LiAl0.05Ni0.95-xCoxO2     | Zhang et al. [260]              | Spray-drying method             | XRD; SEM                   | 153 at 0.2°C      | 40: ~ 6%    |
| LiAl0.05Ni0.95-xCoxO2     | Zhang et al. [261]              | Co-precipitation method          | XRD; SEM; ICP             | 150 at 1°C        | 30: 5%      |
| LiAl0.05Ni0.95-xCoxO2     | Fey et al. [262]                | Co-precipitation method          | XRD; SEM; TEM; XPS; BET    | 207 at 55°C       | 50: 7% at 55°C  |
| LiMn2CoO4                | Wang et al. [256]               | Solid-state reaction at 900°C for 20 h and at 850°C for 15 h | XRD; EPMA; SEM            | 207 at 55°C        | 50: 7% at 55°C  |

**Spinel lithium metal oxides**

| Compound                  | Source                          | Synthesis/Merchandizing Method | Characterization Method(s) | Synthesis temp (°C) | Properties |
|---------------------------|---------------------------------|-------------------------------|-----------------------------|---------------------|------------|
| LiMnO4                    | Pistoia and Rosati [264]        | Solid-state reaction at 730°C for 6 h | SEM                        | ~120                | 80: 16%    |
| LiMnO4                    | Li and Xu [256]                 | Solid-state reaction at 600°C for 6 h and 750°C for 72 h with intermittent grinding | XRD; SEM; EIS          | 112                  | 200: 6.50% |
| LiMn0.9O2                 | Guymard and Tarascon [45]       | Solid-state reaction at 800°C for 24 h | TGA                        | ~ 125               | 200: 12%   |
| LiMn0.9O2                 | Pistoia et al. [240]            | Solid-state reaction at 730°C for 72 h | ~ 100                      | 150: ~ 7%           |
| LiCrMn0.9O4 (p ≤ x ≤ 2)   | Davidson et al. [76]           | Solid-state reaction at 600°C for 3 h and at 100°C for 72 h | XRD; SEM; EIS          | ~ 32 at 0.05°C (x=0.5) | 25: ~ 28%  |
| LiCrMn0.9O4 (p ≤ x ≤ 0.1) | Zhang et al. [52]               | Solid-state reaction at 600°C for 3 h with intermittent grinding followed by slow cooling | TGA; XRD; BET           | 118 (x=0.1)        | 200: 6.70% |
| LiCrMn0.9O4 (p ≤ 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%   |
| LiNi0.9Mn0.1O2             | Wang et al. [75]                | Pechini method                 | XRD; neutron diffraction; EDS | 122(x=0.04)         | 50: 7%     |
| Formula | Authors | Method | Composition | Synthesis Conditions | Properties |
|---------|---------|--------|-------------|----------------------|------------|
| LiM_{1-x}Mn_{x}O (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% |
| LiAl_{1-x}Mn_{x}O (0 ≤ x ≤ 0.6) | 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 |
| LiMn_{1.5}Ni | Myung et al. [26] | Emulsion-drying method | XRD; DSC | 114(x=0.2) | 50: 4% at 45°C |
| LiAl_{1-x}Mn_{x}O (x=0.1, 0.2, 0.3, or 0 ≤ x ≤ 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 x=0.05) 116 (x=0.2 and x=0.07) | 50: ~ 5% 50: ~ 3% |
| LiMn_{1.5}Ni | Amatucci et al. [177,187] | Solid-state reaction at 800°C for 24 h | XRD; SEM | ~100 (x=0.2; x=0.5) | 400: 15% at 55°C |
| LiAl_{1-x}Mn_{x}O (0 ≤ x ≤ 0.3) | 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 |
| LiNi_{0.5}Mn_{1.5}O_{4} | Ogihara et al. | Spray-drying method | XRD; SEM | 112 (x=0.2) | 20: 1.80% |
| Li_{1-x}Mn_{x}O | Yi et al. [273] | Solid-state reaction at 450-750°C for 2 h | XRD; EIS; XPS; SEM; ICP-AES | 124 at 55°C 121 at 55°C | 100: ~ 1% |
| Li_{1-x}Mn_{x}O | Park et al. [271] | Solid-state reaction at 600-900°C for 10 h | XRD; SEM | 126.5 at 25°C 126.5 at 50°C | 100: 0.017%/cycle at 25°C 100: 0.098%/cycle at 50°C |
| Li_{1-x}Mn_{x}O | Patoux et al. [260] | Solid-state reaction at 600-900°C for 10 h | XRD; SEM; BET; AAS | 130 | 500: 8% |
| Li_{1-x}Mn_{x}O | Lu et al. [269] | Solid-state reaction at 450°C for 12 h | XRD; SEM | 80 (x=0) | 50: ~ 1% |
| Li_{1-x}Mn_{x}O | Huang et al. [267] | Solid-state coordination at 700°C for 10 h | XRD; SEM | 104 (x=0.15; y=0.05) | 100: 14.50% |
| Li_{1-x}Mn_{x}O | Shin and Manthiram [270] | Hydroxide precursor method | XRD; EIS; FTIR; TOF-SIMS | 124 at 25°C 121 at 55°C | 100: ~ 1% |
| Li_{1-x}Mn_{x}O | Sun et al. [271] | Solid-state reaction at 650°C for 5 h and 600°C for 10 h | XRD; SEM | 126.1 at 25°C 126.5 at 50°C | 100: 0.017%/cycle at 25°C 100: 0.098%/cycle at 50°C |
| Li_{1-x}Mn_{x}O | Oghihara et al. [250] | Spray pyrolysis | SEM; XRD; BET; AAS | 130 | 500: 8% |
| Li_{1-x}Mn_{x}O | Yang et al. [272] | Urea combustion method | XRD; FTIR; SEM | 133.6 | 20: 0.4% |
| Li_{1-x}Mn_{x}O | Yi et al. [273] | Ultrasonic-assisted sol-gel method | XRD; TGA; TG-TDA; SEM | 126 | 50: 92.1% |
| Li_{1-x}Mn_{x}O | Amaral et al. [274] | Solid-state reaction at 750°C for 72 h | XRD; SEM | 120 (M=Ga and N=S²) | 300: 4% (M=Ga and N=S²) |
| Li_{1-x}Mn_{x}O | 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 LiMn_{1-x}Ni_{x}O | 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_{0.5}O_{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% |
| LiPO_{4}coated LiMn_{0.5}O_{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 LiNi_{0.5}Mn_{0.5}O_{2}powder | Sun et al. [278] | Sol-gel method and coating by in situ mixing for 4 h | XRD; TGA; EDS; TEM; SEM | 137 at 55°C | 50: ~ 0 at 55°C |
| Co-doped LiCo_{0.75}Mn_{0.25}O_{2} (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% |
| LiMn_{3}O_{6} | Huang and Bruce [279] | Modified sol-gel method | XRD | 150 at 0.5°C (synthesized at 200°C) | 300: 40% |
| LiMn_{3}O_{6} | Santiago et al. [47] | Combustion of manganese nitrate tetrahydrate and urea | XRD; EIS | 107 | - |
| LiMn_{2}O_{4} | Yi et al. [46] | Adipic acid-assisted sol-gel method | 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% |
| LiMxMn_{1-x}O_{2} (0 ≤ x ≤ 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% |
| Material system                                                                 | Synthesis method                                                                 | Characterization methods             | Properties                                                                                                                                 |
|---------------------------------------------------------------------------------|-----------------------------------------------------------------------------------|--------------------------------------|-------------------------------------------------------------------------------------------------------------------------------------------|
| **Olivine transition metal phosphates and silicates**                           |                                    |                                      |                                                                                                                                           |
| Mesoporous LiFePO₄                                                              |                                    |                                      | XRD; TEM; BET                                                                  | 155 at 0.1°C; 127 at 1°C                                                                                                               | 30: ~ 0%                                                                 |
| LiFePO₄/graphene/C composite                                                     |                                    |                                      | XRD; SEM; TEM; Mössbauer spectroscopy | 163.7 at 0.1°C; 114 at 5°C                                                                  | 30: 3% (LiFePO₄/graphene2%/C6%)                                                                                               |
| Nanoporous LiFePO₄/C composite                                                  |                                    |                                      | XRD; SEM; TEM; Mössbauer spectroscopy | 165.3                                                                                                                                  | 50: 3%                                                                 |
| LiFePO₄/C                                                                      |                                    |                                      | XRD; TEM; SEM; BET                                                             | 150 at 1°C and 50°C (15 wt% C)                                                            | 30: 14.5%                                                                 |
| LiFePO₄-C/polypyrrole composite                                                 |                                    |                                      | XRD; TEM; BET                                                                  | 128 (5 wt% C)                                                                              | 30: 0.78%                                                                 |
| C-LiFePO₄/polyimide composite                                                   |                                    |                                      | XRD; SEM; TEM; BET                                                             | 160 at 0.5°C (synthesized at 700°C)                                                   | 50: ~ 0% at 0.1°C                                                                 |
| **Carbon-coated LiFePO₄**                                                       |                                    |                                      | XRD; SEM; TEM; SAED                                                            | 156.7 at 0.1°C (synthesized at 850°C)                                                   | 50: 3.5%                                                                 |
| **Carbon-coated LiFePO₄/C**                                                     |                                    |                                      | XRD; SEM; TEM; EIS; EIS                                                       | 150 at 0.05°C; 135 at 1°C                                                                  | 0.1–0.3 mAh/g/cycle                                                                 |
| **Carbon-coated nanocrystalline LiFePO₄**                                       |                                    |                                      | XRD; BET; SEM; XPS                                                            | 122 at 0.1°C and 500°C                                                                    | 85: 11.5%                                                                 |
| **SiO₂-coated LiFePO₄**                                                         |                                    |                                      | 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                                                                                                   |
| **ZrO₂-nanocoated LiFePO₄**                                                      |                                    |                                      | XRD; TEM; SAED                                                                | 146 at 0.1°C                                                                               | 100: 3.1% at 1°C                                                                 |
| **CeO₂-coated LiFePO₄/C**                                                       |                                    |                                      | 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 LiFePO₄/C**                                                          |                                    |                                      | XRD; SEM; EIS                                                                  | 156 at 0.1°C                                                                               | 30: ~ 0%                                                                 |
| **Cu-doped LiFe₉₋ₓCuₓPO₄/C (x=0, 0.01, 0.015, 0.02, or 0.025)**                |                                    |                                      | XRD; SEM; EIS                                                                  | 150 at 0.1°C (x=0.02); 127 at 2°C                                                        | -                                                                 |
| **Co-doped LiFe₉₋ₓCoₓPO₄/F (x=0, 0.03, or 0.06)**                              |                                    |                                      | XRD; XPS; TEM; EDS; Raman spectroscopy | 170 at 0.1°C (x=1/4)                                                                     | -                                                                 |
| **Al-doped LiAlₓV₁₋ₓPO₄/F (x=0, 0.03, or 0.06)**                               |                                    |                                      | XRD; SEM; EIS                                                                  | 118 at x=0.03                                                                               | 30: 14.4%                                                                 |
| **LiFe₉₋ₓTiₓMnx(PO₄)₃/C (0 ≤ x ≤ 0.2)**                                        |                                    |                                      | XRD; SEM; EIS                                                                  | ~118 (x=0.03)                                                                               | 30: ~ 14.4%                                                                 |
| **Li₃Fe₂₋₂ₓTiₓMnx(PO₄)₃/C (0 ≤ x ≤ 0.2)**                                      |                                    |                                      | XRD; SEM; EIS                                                                  | 112.3 (x=0.1)                                                                               | 20: 25.8%                                                                 |
| **LiFePO₄ powders**                                                             |                                    |                                      | 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                                                                                           |
| **LiFePO₄/C nanocomposites**                                                    |                                    |                                      | XRD; SEM; TEM; EDS; Raman spectroscopy | 141 at 0.2°C                                                                              | -                                                                 |
| **LiCoPO₄/C**                                                                  |                                    |                                      | XRD; SEM; TEM; EIS                                                            | 136.2                                                                                      | 30: 32%                                                                 |
| **LiCoPO₄/C nanocomposites**                                                    |                                    |                                      | XRD; SEM; TEM; EIS                                                            | 109 at 0.05°C; 142 at 20°C                                                               | 40: 13% at 0.1°C                                                                 |
| **Vanadium-based compounds**                                                    |                                    |                                      | XRD; SEM; TEM; EIS                                                            | -                                                                                          | -                                                                 |
| **LiV₃O₈**                                                                      |                                    |                                      | XRD; TGA                                                                     | -                                                                                          | 50: 0.5%/cycle                                                                 |

**References:**
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| Material                  | Authors (Year) | Synthesis Method                  | Characterization Methods | Temperature | Capacity (%) |
|--------------------------|----------------|------------------------------------|--------------------------|-------------|--------------|
| Li6V10O28                | Xie et al. [151]| Hydrothermal synthesis and annealing dehydration treatment | XRD; TGA; TG-TDA; ICP-AES; SEM; TEM | 132         | 15: ~ 24%    |
| LiV2O5                  | Xiong et al. [120] | Spray-drying method | XRD; SEM | 340.2 at 25 mAg⁻¹ | 100: 15.2% at 125 mAg⁻¹ |
| LiV2O5                  | Karan and Manthiram [105] | Low-temperature solution dispersion method | XRD; SEM; DSC; TGA | ~ 250 at 60°C | 100: ~ -4%   |
| Li2V4(PO4)3/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%   |
| Li2V4(PO4)3/C           | Wang et al. [140] | Electrostatic spray deposition | XRD; SEM; TEM | 127.9 at 1°C (synthesized at 750°C) | 100: ~ 0% at 1°C and 5°C |
| Li2V4(PO4)3/C           | Yuan et al. [301] | Sol-gel method | XRD; SEM | 122 at 0.5°C at -25°C | 100: 5% at 20°C |
| Cr-doped LiV2O4         | Feng et al. [122] | Sol-gel method | XRD; SEM | 269.9 at 150 mAg⁻¹ | 100: 5.6% |
| Polypyrrole-LiV2O4 composite | Feng et al. [123] | Solution dispersion in ethanol followed by co-heating process | SEM | 292 at 40 mAg⁻¹ (20% PPy) | 40: ~ 14 |
| Mixed amorphous-nanocrystalline LiV2O4 | Shi et al. [127] | RF magnetron sputtering | XRD; TEM; SEM; SAED | 382         | 100: 21.2%   |
| LiV2O4/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%   |
| LiV2O4 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 LiV2O4       | Ma et al. [126] | Tartaric acid-assisted sol-gel process | XRD; SEM; TEM | 301 at 40 mAg⁻¹ | 50: 3.7% |
| Polypyrrole-LiV2O4 composite | Tian et al. [125] | Oxidative poly-merization of pyrrole using ferric chloride | XRD; FTIR; SEM | 169 at 0.5°C | 50: ~ 0% |
| LiV2O4/C nanorods       | Xu et al. [128] | Hydrothermal synthesis | XRD; TGA; TEM; FTIR | 302 at 300°C | 30: ~ 8% |
| LiV2O4 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 Fe2V4O13      | Si et al. [128] | Liquid precipitation method | XRD; SEM | 235         | 40: ~ 14.5% |
| Mn0.02V2O5 (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**

| Material                | Authors (Year) | Synthesis Method                  | Characterization Methods | Temperature | Capacity (%) |
|-------------------------|----------------|------------------------------------|--------------------------|-------------|--------------|
| K2FeO4                  | Wang et al. [304] | Hypochlorite oxidation method | XRD; CCD | ~ 350 | 50: ~ 35% |
| LiFePO4 powder          | Oghihara et al. [250] | Spray pyrolysis | SEM; XRD; BET; AAS | 150 | 500: 16% |
| LiFe/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) |
| LiMn2O4/CNT nanocomposites | Ding et al. [73] | In-situ hydrothermal method | XRD; SEM | 116 at 1°C 84 at 10°C | 100: 0.009% at 1°C 100: 5.6% at 10°C |
| LiMn2O4/CNT nanocomposites | Xia et al. [307] | In-situ hydrothermal method | XRD; TEM; SEM; TGA | 124 at 1°C 106 at 10°C | 500: 8% at 1°C 1000: 23% at 10°C |
| LiFePO4/multiwalled CNT composites | 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 | 155 at 0.1°C 147 at 1°C | 50: ~ 5% at 0.1°C 50: ~6% at 1°C |
| Li[Li1/3Mn2/3]Ni0.2Co0.15O2-V2O5 composites | Gao et al. [119] | Co-precipitation method | XRD | 300 at 0.05°C (10 wt% V2O5) | 25: ~ 12% |
| V2O5/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% |
| Advanced Cathode material | PTMA | Nakahara et al. [171] | radially polymerizing | SEM, - | 1000: 11% |
|----------------------------|------|----------------------|---------------------|-------|-----------|
| TCNO/DOQ/THBQ | Hanyu and Honna [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 | Marmorstien et al. [173] | Mixing powder | Electrochemical characterization | 1600 at 90-100°C | 10: 30% |
| LiS | Yang et al. [174] | Ball milled | SEM | 800 | 10: 0.25% |

Conversion Cathodes

Metal Fluoride

FeF/C, CMFNCs | Badway et al. | High energy mechanical milling | XRD, transmission electron microscopy | 200 600 at 70°C | 50: 10% |
|----------------|----------------|---------------------------------|---------------------------------|----------------|-----------|
| FeF/C | Li et al. [175] | Ball milling | XRD; TEM | 712 | 10: 16% |
| (MF3) FeF3, TiF4, and VF5 | Arai et al. [178] | Lithiated; delithiated | XRD | 140 at 25°C | 35% |
| LiInI3O2F | Kubo et al. [179] | Solid state reaction | XRD | 240 | 30:- |
| LiInMo2O4F | Choi and Manthiram [180] | Solid-start reaction | XRD | 150 at 500 and 600°C | 50:- |

Carbon Fluoride

FeF3, FeF2, and BiF3 | Amatucci et al. [185] | solid state prelithiation process resulted | XRD, DMC, and TEM | 243 | 0.18 at 24°C and |
|-----------------|---------------------|---------------------------------|---------------------------------|----------------|-----------|
| (CF)n/(C2F)n | Watanabe [186] | Mild reaction conditions | XRD;ESCA spectra; DTA | - | - |
| IF4/ HF | Yazami [187] | RT and HT | XRD; IR spectroscopy | - | - |
| Graphite | Wang et al. [188] | graphene paper prepared chemically with graphite | SEM | 298 680 528 | 50: 19% 2: 87% |
| LIBF3 | Yazami et al. [190] | Solid state | EPR; XRD; Raman spectroscopy; TEM | 865 | - |
| IF3, NbF5, MoF6, WF6, ReF6, BF3 | Hamwi et al. [191] | prepared by reaction of IF 5 with excess fluorine at 300°C | XRD; IR spectroscopy studies | - | - |

Fluorophosphates

LiFePO4/LiFe2O3/PO2/PO3 | Barker et al. [1] | Novel carbothermal reduction (CTR) | XRD | 150 | - |
|----------------|-----------------|---------------------------------|----------------|----------------|-----------|
| 1. LiP2O7 | Lin et al. [199] | sol-gel, calcinations | XRD;TEM | 130 | 30: 4.6% |
| LiP2O7F/C | Wang et al. [200] | Annealing | XPS; TG-DTA;XRD | 136 at 0.1°C rate 123 at 1°C rate. | 100: 4.7% |
| LiP2O7F/LiCoO2 | Gover et al. [196] | Hydrothermal method | SEM; XRD; TEM | 143 | 300 at 45°C 280 at 60°C |
| LiP2O7/C | Zhang et al. [201] | Hydrothermal method | SEM; XRD; TEM | 143 | 50: 2.8% |
| Na3V2(PO4)2F3/VPO4 and NaF | Gover et al. [202] | Solid state carbothermal reduction | XRD | 120 | 220:- |
| Na3V2(PO4)2F3/NaF/LiC104 | Gover et al. [203] | Hydrothermal method | XRD; SEM | 117.3 and 106.8 | 100: 1.3% |
| Na3V2(PO4)2F3 | Jiang et al. [204] | Sol-gel method; heat treatment | TGA; TEM | 127 and 117 | 30:- |
| Na3V2(PO4)2F3 | Song et al. [205] | Sol-gel method; heat treatment | XRD | 188 | 37: 3.2% |
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 LiPF₆, 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 principle, the layered lithium transition metal oxide with the formula LiMO₂, 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).

**Cathode materials**

Despite the fact that layered LiCoO₂ 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 LiNi₀.₅Mn₀.₅O₂ and LiMnO₂ along with their derivatives such as LiNi₀.₅Mn₀.₅O₂, LiCoNiₐMn₁₋ₐO₂, spinel-structured LiMnO₂ along with its derivatives such as LiNiₐMn₁₋ₐO₂ and LiCrₐMn₁₋ₐO₂, and olivine-structured LiFePO₄. 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 LiMO₂, 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).

**LiCoO₂**

As stated earlier, LiCoO₂ 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 LiCoO₂. This was further motivated by the high cost, chemical hazards, and the environmental impact associated with cobalt. The preparation of LiCoO₂ 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...
Structural change

Insertion

“Usual” Processes

Commercialized

Very large capacity

Volume changes are still an issue

Under development

Large capacity

Voltage hysteresis

Figure 2: Structural changes during charge-discharge of Insertion, alloying and conversion cathodes materials.

Figure 3: Voltage and specific capacity of different cathode materials.

[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, LiCoO₂ could have either hexagonal layered for high-temperature LiCoO₂ or cubic spinel-like structure for low-temperature LiCoO₂ [23]. A comprehensive review of high- and low-temperature synthetic methods and their effects on the electrochemical properties of LiCoO₂ was conducted by Antolini [23].

Despite its high theoretical capacity of 274 mAh⁻¹, reported practical discharge capacities of LiCoO₂ are relatively low, in the range of 135-150 mAh⁻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 Al₂O₃, ZnO, and LiTiO₂, 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 LiCoO₂ cathodes have been studied including AlPO₄, Al₂O₃ [29,30], CePO₄, FePO₄ [16], LiCoO₂, Li₃PO₄N [31], Li₃TiO₄ [32], MgO [33], SrHPO₄, SrO [34], SnO₂, TiO₂ [34,35], ZnO [36], and ZrO₂ [35,37]. Having achieved a mixed record of success, some coatings remarkably increased the initial discharge capacity of LiCoO₂ as high as 190 mAh⁻¹ while others significantly enhanced the cycling behavior. For instance, Lu et al. [11] reported an initial discharge capacity of 195 mAh⁻¹ for Al₂O₃-coated LiCoO₂ (1.0 wt% Al₂O₃) 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% FePO₄-coated LiCoO₂ cathodes. Other coatings which have produced significant improvements on the electrochemical behavior of LiCoO₂ include ZnO [38,39], ZrO₂ [35], Li₃PO₄N [31], and Li₃TiO₄ [32].

LiMnO₂

Although LiMnO₂ has been proposed as a cathode material in LIBs almost as early as LiCoO₂, 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 LiCoO₂, LiMnO₂ 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 (LiNiO₂) which has a comparable layered structure and
charge–discharge characteristics to those of LiCoO₂. 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 LiNiO₂ 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), whereas 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 LiNi₃CoO₄, LiCoO₂, LiNiM₂O₄, and LiNi₃M₃O₄ (where M=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 LiNi₃Co₂Ti₅O₁₄M₈O₄ (≈ 0% after 20 cycles) [41].

**Spinel Lithium Transition Metal Oxides (LiM₂O₄)**

LiMn₂O₄

The spinel lithium manganese oxide LiMn₂O₄ 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 LiMn₂O₄ 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 LiMn₂O₄ reported a discharge capacity in the range of 100–120 mAh/g which represent 67–81% of its theoretical capacity (148 mAh/g) [48]. A wide variety of synthetic approaches have been applied to develop spinel LiMn₂O₄ 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 LiM₃Mn₂-xO₄ (M=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 LiMn₂O₄ [52–61]. Among these derivatives, LiNi₃M₉O₂ showed significant improvement on cycling behavior of LiMn₂O₄. For example, Wang and Xiao et al. showed a relatively good cycling behavior and higher reversible capacity of LiNi₃M₉O₂ 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 LiMn₂O₄. The theoretical capacities of 100–120 mAh/g, slightly lower than that for LiMn₂O₄ [63].

Surface modification has been a second way to improve the electrochemical performance of LiMn₂O₄. Different salts have been studied such as Al₂O₃, AlPO₃, Cr₂O₃ [64], La₂O₃ [65,66], LiPO₄ [67], SrF₂ [68], TiO₂ [69], ZnO [70,71], and ZrO₂ [69]. Further nanostructural modifications have been examined such as encapsulation of LiMn₂O₄ nanowires in ZnO nanotubes by Liu et al. [72] and homogenous dispersion of LiMn₂O₄ nanoparticles in carbon nanotubes (CNT) composites by Ding et al. [73]. The latter approach demonstrated a significant improvement on the cycling behavior of LiMn₂O₄ approaching 100% after 100 cycles [73].

**Derivative Compounds**

Much research work has been conducted on the electrochemical properties of derivatives of LiMn₂O₄ 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, LiNi₄Mn₂O₄ [74], and LiCr₄Mn₂O₄ [52,75,76], have demonstrated a remarkable improvement on the cyclability of spinel magnesium oxides.

**Olivine Lithium Transition Metal Phosphates and Silicates (LiMPO₄ and LiMSiO₄)**

LiMPO₄

The electrochemical behavior of olivine-structured transition poly anion compounds of the structural formulas of LiMPO₄ and LiMSiO₄ (M=Co, Fe, Mn, Ni or V) have been attributed 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 a 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 LiMnSiO 4 [87-89], Li2CoSiO4 [90-92], Li2FeSiO4 [78,88,93-102], and LiFeSiO 4 [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 V 2O 5 [106], V 3O 4 [104,106-117] V 3O 4 [118], V 4O 9·H 2O [119-125], LiV 3O 8 [104,126-139], Li V4(PO) 4 [140-146], V 5O 8 [147], Fe 2V 4O 13 [148], V 6O 13 [149,150], and LiV 3O 8 [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 Li V4(PO) 4 , have shown the most promising electrochemical behavior as indicated by their high discharge capacities and good capacity retention. Li V4(PO) 4 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 250-300 mAhg⁻¹ for V 2O 5/polypropylene 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 Li V 2O 5 (x~0.8) nanorods with thickness of 5-10 μm 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 mAh g\(^{-1}\) with low capacity loss (~15% after 100 cycles) for Li\(_2\)O\(_4\)/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 mAh g\(^{-1}\)) with good cycling efficiency (8-14% capacity loss after 30-40 cycles) for polypyrrole-Li\(_2\)O\(_4\) 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 LiMn\(_2\)O\(_4\) include SiO\(_2\) [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 LiFePO\(_4\)/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(2,6-terthathenyl-1-piperidinolxy-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 Li\(_2\)S**

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]. Li\(_2\)S 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 Li\(_2\)S 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 (FeF\(_3\)) 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 fluoride-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\(_{2}\)), spinel lithium manganese oxyfluorides (Li\(_{10}\)Mn\(_{2}\)O\(_4\)) and orthorhombic lithium manganese oxyfluorides (Li\(_{10}\)Mn\(_{13}\)O\(_{12}\)F\(_{10}\)) [187].

**Layered transition metal oxyfluoride:** As have been mentioned before about layered 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\(_{2}\) F:** Kubo et al. [180] discussed metal oxyfluoride as cathode materials by studying the synthesized of Li\(_{1-x}\)Ni\(_{1-x}\)O\(_{2}\) F\(_{y}\) through solid state reaction that led to substitute nickel and oxygen positions for LiNiO\(_{2}\). X-ray diffractometry and electrochemical technology were used to examine the cycle properties and show initial charging capacity around 240 mAh/g. Li\(_{1-x}\)Ni\(_{1-x}\)O\(_{2}\) F\(_{y}\) possessed
excellent features on charge/discharge cycling compared to LiNiO₂ and showed enhanced in the capacity retention during cycling [188-190].

**Spinel lithium manganese oxyfluorides:** Spinel lithium manganese oxide LiMn₂O₄ has been studied widely as cathode materials due to its features of being non-toxic, inexpensive and environmentally-friendly material. Spinel LiMn₂O₄ can deliver 120 mAh/g capacity. Choi and Manthiram [182] reported the synthesized of spinel Li₄Mn₅O₁₂ Fₙ oxyfluoride cathodes at 500 and 600°C by substituted F-ions from employing LiF for oxygen (O₂⁻) ions in the spinel Li₄Mn₅O₁₂ Fₙ XRD, electrochemical cycling, and chemical analysis measurements were carried out to characterize Li₄Mn₅O₁₂ Fₙ 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 Li₂O, 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]. VF₃ and TiF₃ 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 fluorides with metal fluorides by prepared pre-lithiation agent, Li±.N, to (FeF₃, FeF₄, and BF₄) metal fluorides. XRD, DMC, and TEM measurements were carried out to characterize the structure of the nanocomposite product and showed 243mA/hg 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, (CF₂), and (C, F), respectively while LT fabricated fluorine-graphite intercalation compounds of CF₆ [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 Wh 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. LiMPO₄F and Li₂MPO₄FM (M=Fe, Ni, 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, LiVPO₄F, NaVPO₄F, and LiFePO₄F [63].

**LiVPO₄F:** 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 LiVPO₄F 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]. Cover et al. [210] prepared LiVPO₄F as a cathode electrode in LIB using carbothermal reduction method, DSC, and electrochemical measurements were performed. The discharge capacity is about 140 mAh g⁻¹ for the positive electrode with average discharge voltage around 4V [200]. Other research done by Zhang et al. [208] depended on prepared LiVPO₄F/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].

**NaVPO₄F:** Sodium vanadium fluorophosphate characterized as a safer, economical and higher work potential comparing to other materials [209,210]. This includes Na₅V₃(PO₄)₃Fₙ with theoretical capacity equal to 128 mAh/g [170,211,212], NaVPO₄F [209] and Na₃V₂(PO₄)₂F₃ [213]. Generally, NaVPO₄F materials synthesis required VPO₄ 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]. Na₅V₃(PO₄)₃F_n 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 Na₃V₂(PO₄)₂F₃ delivered through 3.6 and 4 V vs. Na⁺/Na theoretical capacity equal to 156 mAh/g [217-221].

**LiFePO₄F:** Another option about olivine-type, variety-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]. LiFePO₄F was conducted by Ramesh et al. [218] as a new adequate material of fluorophosphate because of Li⁺ feature that migrates easily without any barriers. LiFePO₄F 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] LiFePO₄F 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 LiFePO₄. Other various fluorophosphate materials have been studied including LiFePO₄F [218,226-230], Li₃MPO₄F (M=Co, Ni) [231-235], and Na₅V₂(PO₄)₃F Hybrid Ion Cathode [204,210].

**Fluorosulfates**

New material of cathode materials reported first by Sebastian et al. was LiMgSO₄F, they presented the benefit of LiMgSO₄F (M=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 variety type structure LiFePO4 OH. Investing the large electronegativity of sulfur and fluorine of LiMSO4F in produce high-voltage cathode materials such as LiFeSO4F 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].

LiFeSO4F: Ionothermal was the first method to produce LiFeSO4F, it can be reused repeatedly for synthesizing although it is very high cost ionic liquid media [245-250]. LiFeSO4F 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 variety-type LiFeSO4F has low thermodynamic stability, unstable at temperatures above 350°C, because of that, producing single-phase LiFeSO4F It is difficult to occur [103,256-259]. LiFeSO4F has a theoretical capacity of 148 mAh/g and produced an energy density (543 Wh/kg) as good as LiFePO4 (581 Wh/kg) [260-263]. Rosati et al. [264] reported the reversible capacity of approximately 85 mAh g-1 of LiFeSO4F which prepared in the trilite structure by the ceramic method at 300°C [265-269]. LiFeSO4F has numerous advantages as cathode material such as it can be prepared from plentiful FeSO4·H2O precursor and synthesized at low temperature [270-276].

LiMSO4F (M=Co, Ni, Mn, Zn): Fluorosulfate polyanionic LiMSO4F electrode materials represent a wide family of a good combination of properties, especially, both electrochemical and safety issues. LiMSO4F showed a redox potential of 4.25, 4.95, and 5.25 V, respectively [277-281]. LiCoPO4 is considered as a 4.9 V while LiNiPO4 is considered as a 5.4-V cathode [231,281-285]. In contrast with Fe 2+/Fe3+ redox reaction, Ni2+/Ni3+ redox reaction, Mn2+/Mn3+ and Co3+/Co2+ redox reactions do not take place within the explored galvanostatic cycling potential window [286-291].

LiMSO4F (M=Co, Ni, Mn, Zn) synthesized at low temperature similarly to LiFeSO4F by using solid-state [291-296] and polymer-assisted methods [297-300]. Various other fluorosulfates materials have been studied including LiFe1-xMxPO4F (M=Mn, Zn), NaMSO4F (M=Fe, Co, Ni, Mn, Mg, Zn, Cu), NaMSO4F·H2O (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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