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Metal oxides, metal sulphides and hybrid cathode materials for aluminium ion batteries – a mini review

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Abstract. Immense progress is required in the field of energy conversion and storage to achieve cost-effective high-performance batteries which could meet future energy demands. It also becomes mandatory to utilize the abundant materials from the earth's crust, along with safe conversion and storage of electrochemical reactions. In this perspective, aluminium ion batteries (AIBs) could be a viable alternative for the conventional lithium-ion batteries (LIBs). In this mini-review, we focus on the challenges and the latest growth related to the cathode active materials (metal oxides, metal sulfides and other hybrid systems) in AIBs. The development of highly efficient, low cost and safe energy storage systems based on aluminium ion batteries is much explored. Still, commercialization remains a theoretical one, hence this review highlights the significance of the developed materials and their shortcomings that need to be addressed for the commercialization of aluminium ion batteries in the future.

1. Introduction

The lithium-ion technology, with its high specific energy and power density, is the most extensively used energy storage systems in portable electronic devices over the past 20 years [1- 3]. Although LIBs have dominated the consumer electronics market, the unavailability of lithium, flammability and high cost lead to the transformation of the prevailing research focus towards an alternative technology [3, 4]. To accomplish long-term sustainability, lightweight, toxicity and high cost the exploration of an alternative energy storage system can be drifted to materials like aluminium, magnesium, sodium and potassium. Special attention is focussed on AIBs owing to the abundance of Al, low reactivity, easy handling with a high volumetric capacity of 8046 mAh cm⁻³ [4] and gravimetric capacity of 2980 mAh g⁻¹. Figure 1 compares the volumetric and gravimetric capacity of various (Al, Li, Mg, Ca, Zn, Na, K) battery systems. From the comparison, it can be ascertained that AIBs have superior capacity compared to the commercial LIBs and other battery systems.

In fact, rechargeable aluminium ion batteries are considered as a notable candidate for the future energy demands. Since the three electron transfer during the electrochemical charge-discharge process in AIBs provides an efficient storage capacity than LIBs [5]. Added to this the fabrication of electrochemical cell using aluminium can be done in an open environment which uplifts the safety of



aluminium ion battery systems [6]. All of the mentioned batteries operates with the basic principle of intercalation/de-intercalation mechanism, where the cathode delivers the positive potential and the anode generates the cell voltage [7]. The first voltaic cell proposed by Sargent, employs Al anode, carbon as a cathode and aqueous caustic alkaline solution with dissolved zinc oxide [8]. The first secondary aluminium battery was reported by Holleck and Giner with the use of chlorine as a cathode, Al metal anode in an electrolyte composed of AlCl_3 , KCl , NaCl [9].

Many improvements in the anode and cathode material, electrolytes and additives were investigated and numerous novel results have also emerged [7]. Progress in the cathode materials was stimulated in the last decade and several materials have been assessed to obtain AIBs with good efficiency, safety and low cost. Based on the results obtained with the development of unique materials and enhanced volumetric and gravimetric capacity relative to LIB systems, AIBs could be the future generation energy storage systems. However, the commercialization and development of AIBs are hindered by lack of efficient cathode materials to produce batteries with high specific capacity and stable cycle life [1, 7]. Several cathode materials have been reported so far, despite that still the commercialization of AIBs remains underdeveloped. The foremost reason being the complex issues associated with the cathode materials such as low discharge voltage [11, 12], poor cycle life [5, 13], low coulombic efficiency, unstable discharge capacity, volume expansion [14, 15] and unstable cycling ability [15].

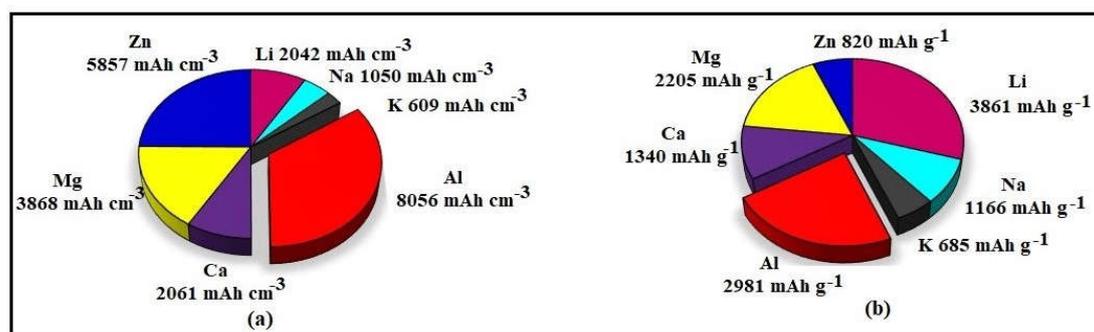


Figure 1. Comparison of the volumetric and gravimetric capacity of Al, Li, Mg, Ca, Zn, Na, K battery systems.

This paper is focused on the recent development of the cathode materials based on metal oxides, metal sulphides and hybrid materials with carbon, graphene, (carbon nanotubes) CNTs for AIBs. The cathode materials based on graphite is not discussed in this review since it is a vast topic and entails an extravagant study. In order to have a profound understanding of the disputes associated with the current cathode materials and suggested requirements of the future cathode materials for successful commercialization will also be addressed.

2. Cathode materials

The cathode materials for AIBs were developed by taking into consideration the materials employed in LIBs since the mechanism involved in both the cases are similar. The cathode active material in the battery system is one of the key parameters in building highly efficient battery systems with high power and energy density for commercial applications [7, 17]. The development of AIBs is hampered by the non-availability of cathode materials for practical applications and still, the outline and the principles of cathode materials remains un-determined [1]. Over the past decade, various cathode materials were proposed for AIBs which is discussed in detail.

2.1. Metal oxide-based cathode materials

Wang et al in 2013, for the first time, attempted the use of vanadium oxides as electrode materials for aluminium ion batteries. The tunnel structure of VO_2 and the suitable electrochemical potential window

lead to its use in batteries as a cathode material. The VO₂ nanorods prepared by hydrothermal treatment has a specific area of about 37.3 m² g⁻¹, responsible for shorter diffusion length. The cyclic voltammetry studies were performed in the potential ranging from 0.0 to 0.9 V and at a current density of 50 mA g⁻¹, the discharge capacity of 165 mAh g⁻¹ is achieved for an initial cycle which is declined to 116 mAh g⁻¹ after 100 cycles. This is the initial work based on aluminium ion batteries sustaining the capacity for 100 cycles [17]. Hence, Wang et al. initiated the use of metal oxides as an electrode material in aluminium ion batteries which led to several other developments on metal oxides.

Other oxides of vanadium i.e., V₂O₅ is also considered to be a flexible host material for batteries which works on the principle of intercalation/de-intercalation mechanism such as lithium, sodium, aluminium and magnesium battery systems [18]. The reason for its use in batteries is the layered structure of V₂O₅, water absorption ability and high redox potential of V⁵⁺. The electrochemical behavior of V₂O₅ is influenced by the intercalated water which has a profound repercussion in AIBs relative to the other battery systems [5, 20]. Gonzalez et al., in 2016, reported the electrochemical nature of V₂O₅ xerogel in aqueous AlCl₃ electrolyte using Swagelok type cells. The smaller water content present in V₂O₅ camouflaged the repulsion occurring between oxygen within the Al_xV₂O₅ during the intercalation process. The chemically intercalated aluminium ions with positive charge balance the negative charge on vanadium oxide, the results also suggest that V₂O₅ may suffer from limited intercalation with subsequent release of oxygen. It was also reiterated that there is no possibility for electrolyte decomposition and corrosion in the prescribed voltage range using V₂O₅ as an electrode material [19].

With all these different oxides of vanadium being reported as one of the efficient cathode material for aluminium ion batteries still, the controversy exists regarding the contribution of V₂O₅ to the capacity of those batteries [20]. This leads to a comprehensive investigation of the storage mechanism of Al³⁺ ions using metal oxide based electrode materials. This was addressed by Gu et al., in 2017, by preparing V₂O₅ nanowire with a diameter of about 80-120 nm by hydrothermal treatment [21]. To validate that the capacity obtained using V₂O₅ electrode doesn't arise from the current collector, Galvanostatic cycling was performed on bare nickel foam as a current collector without the addition of V₂O₅, which unveils zero capacity [10]. The specific capacity achieved using V₂O₅ nanowire is 107 mAh g⁻¹, quite low compared to the other reported results in V₂O₅ [21]. A two phase-transition reaction and intercalation of Al³⁺ ions into the metal oxide with the formation of amorphous layers was observed. This study provides a new pathway and detailed manifestation, substantiating the participation of V₂O₅ in the electrochemical reaction which gives rise to the obtained capacity.

The anatase form of TiO₂ being nontoxic and chemically stable is widely used in several energy storage devices [23, 24] and self-cleaning applications [25, 26]. Liu et al in 2012 synthesized TiO₂ nanotube array film with a thickness of about 14 μm by anodizing the metallic titanium foil. The nanotube array promotes an exceptional contact between the electrode and the electrolyte along with diminished ion diffusion with high electronic conduction. The intercalation process is highly dependent on the anion from the electrolyte accompanying Al³⁺ ions. The discharge capacity of 75 mAh g⁻¹ was acquired at a current density of 4 mA cm⁻² and TiO₂ nanotube offers a boosted discharge capacity compared to the anatase TiO₂. Another reason for the improved performance is the nanotube structure, facilitating fast transport of electrons and a short ion diffusion path. The aluminium storage is calculated from the highest discharge capacity and it was observed to be 0.074 mol per anatase. The reason beyond the reported discharge capacity is owing to the reversible electrochemical redox reaction of Ti⁴⁺/Ti³⁺ during the intercalation process [12]. But TiO₂ nanotube suffers from the formation of oxide film on the Al anode surface and corrosion, hampering the redox reactions which lowers the cell potential [6].

Based on the previous reports on TiO₂ as an electrode material for LIBs, He et al, fabricated black anatase phase TiO₂ nano leaves for rechargeable aluminium ion battery and compared the results with the commercial white anatase phase TiO₂. The discharge capacity of commercial TiO₂ was 77.2 mAh g⁻¹ and for the TiO₂ nano leaves initial discharge capacity of 278.1 mAh g⁻¹ was reported, which is approximately 4 folds higher than the former. The irreversible capacity loss in TiO₂ nano leaves is only 2.84 % with excellent reproducibility and smaller capacity fading even after 300 cycles. The reason behind this superior performance of nano leaves is its shorter diffusion length, larger interfacial area and

lower polarization. It is noteworthy to mention that the discharge capacity and the cycling stability obtained in this work is far superior when compared to other TiO₂ reports available for AIBs [5]. The summary of metal oxide based cathode materials and its initial discharge capacity is tabulated in table 1. Only a few reports are available on the development of metal oxide based electrode materials for AIBs unlike LIBs, since in the reported work its cell voltage, efficiency and cycle life were not much pronounced even though they show a higher capacity. Hence much focus is directed towards metal sulfides and other hybrid systems.

Table 1. Summary of metal oxide based cathode materials and its initial discharge capacity.

S. No.	Cathode Material	Electrolyte	Discharge capacity (mAh g ⁻¹)	Ref.
1.	V ₂ O ₅ nanowire	AlCl ₃ + [EMIM]Cl	305	[21]
2.	VO ₂	AlCl ₃ + [BMIM]Cl	116	[17]
3.	V ₂ O ₅ xerogel	AlCl ₃ + deionized water	130-250	[19]
4.	TiO ₂	Aqueous AlCl ₃	75	[12]
5.	TiO ₂ nanoleaves	Al(NO ₃) ₃	278	[5]
6.	Commercial TiO ₂	Al(NO ₃) ₃	77	[5]

[EMIM]Cl = 1-ethyl-3-methylimidazoliumchloride, [BMIM]Cl = 1-butyl-3-methylimidazoliumchloride

2.2. Metal sulfides based cathode materials

Among transitional metal sulphides, NiS is considered as an efficient electrode material for lithium ion batteries because of its good conductivity, theoretically high capacity and excellent catalytic activities [26]. Related to its use in lithium-ion batteries, hexagonal NiS nanobelts with length around 10-25 μm and size of 50–100 nm were synthesized by hydrothermal method and used as an electrode material for aluminium ion batteries. Pouch cells were assembled using NiS nanobelts as a cathode and high purity Al as an anode, the discharge capacity of 104.7 mAh g⁻¹ was attained for the first cycle, NiS with nanobelt structure eases Al³⁺ ions diffusion. With further increase in the cycles, discharge capacity increases and at 100th cycle the discharge capacity of 104.4 mAh g⁻¹ was accomplished at a current density of 200 mAh g⁻¹. Although, the synthesized NiS nanobelts was able to retain 90% capacity over 100 cycles it endeavors from low cell voltage hindering its use for the practical system [27]. Advancement is made by means of integrating graphene with Ni₃S₂ in 2016 by Wang et al. The discharge capacity of 295 mAh g⁻¹ which slowly decreased for consecutive cycles with a coulombic efficiency of 84.3%. Although the coulombic efficiency is low, there exists an enhanced reversible charging and discharging process. The integration of Ni₃S₂ sheets with graphene matrix can be reaffirmed by the low internal resistance of about 10Ω [28].

TiS₂ and cubic Cu_{0.31}Ti₂S₄ are also encountered as cathode materials for AIBs. The electrochemical intercalation of Al in titanium sulfides as the electrode was experimented at both room temperature and at 50°C since in the latter temperature an advanced capacity and distinctive charge-discharge characteristics were observed. The total capacity of TiS₂ with a layered structure is 70 mAh g⁻¹, which was reported to be gradually uplift upon the consecutive cycles. Although the behavior of TiS₂ in room temperature and 50°C are similar, the capacity is more at higher temperature due to the better reaction kinetics. The author also hypothesized that with both the sulfides based cathode high capacity was not accomplished because of the strong coulombic attraction between aluminium and sulphide ions [29]. Hence, the future studies should focus on improving Al³⁺ ions transport by doping of ions in the crystal structures.

A new class of electrode material which is worth mentioning is based on chevrel phase molybdenum sulphide (Mo₆S₈), used in rechargeable magnesium-ion batteries as a cathode material for the first time by Aurbach and co-workers [30]. The unique structure of Mo₆S₈ is utilized initially by Linxiao et al. in

2015 as an electrode material for aluminium ion battery using $\text{AlCl}_3\text{[BMIM]Cl}$ electrolyte. The discharge capacity of 148 mAh g^{-1} , declined to 70 mAh g^{-1} for the 2nd cycle and stabilized after 50 cycles exhibiting promising cycle stability [31]. The low specific capacity may be due to the high molecular weight of Mo_6S_8 . Recently, MoS_2 a chalcogenide from the transition metal family with microspherical morphology was synthesized by hydrothermal route and employed as a cathode material in AIBs. The discharge specific capacity of 253.6 mAh g^{-1} was reported for a current density of 20 mA g^{-1} and the charge is stored between the electrode and electrolyte interface [32]. Further, the detailed mechanism for the intercalation and de-intercalation of Al^{3+} ions to the electrode material during the charging/discharging process was elaborated in detail with the crystal structure. The coulombic efficiency of MoS_2 as cathode material is low, the reason being the phase transition of MoS_2 and formation of solid electrolyte interface (SEI) on the surface of the electrode [33]. Very recently $\text{Al}_{4/3}\text{Mo}_6\text{S}_8$ was developed by Agiorgousis et al., which could function as a promising material for energy generation and storage, i.e., act as a Janus material (dual function) [34].

Iron disulphide has gained consideration in LIBs as a cathode active material because of its high theoretical capacity and non-toxic nature [35] and hence it has been used extensively in lithium and sodium batteries [36]. In terms of sulfides, very recently the first attempt of employing FeS_2 as cathode active material was exemplified by Koura et al., operating aluminium rechargeable battery systems at a low temperature of about 25°C and an elevated temperature of about 240°C respectively [37]. Based on these studies FeS_2 was used as electrode material for rechargeable aluminium batteries operating at 55°C and the possible charge/discharge reaction mechanism was illustrated by Mori et al in 2016. The theoretical capacity of FeS_2 as an electrode material is 447 mAh g^{-1} and in this reported study about 85% was achieved, the smaller loss in the capacity may originate from the unreacted FeS_2 in the electrochemical cell. It is also suggested that a phase transition from FeS to FeS_2 phase may occur during the charging process [11]. The summary of metal sulfide based cathode material and its initial discharge capacity is given in table 2.

Table 2. Summary of metal sulfide-based cathode materials and its initial discharge capacity.

S. No.	Cathode Material	Electrolyte	Discharge capacity (mAh g^{-1})	Ref.
1.	NiS nanobelts	$\text{AlCl}_3 + \text{[EMIM]Cl}$	105	[27]
2.	TiS ₂	$\text{AlCl}_3 + \text{[BMIM]Cl}$	70	[29]
3.	Cu 0.31Ti ₂ S ₄	$\text{AlCl}_3 + \text{[BMIM]Cl}$	25	[29]
4.	Mo ₆ S ₈	$\text{AlCl}_3 + \text{[BMIM]Cl}$	148	[31]
5.	MoS ₂	$\text{AlCl}_3 + \text{deionized water}$	254	[33]
6.	FeS ₂	Aqueous AlCl_3	390	[11]

2.3. Other hybrid systems

When the electrode material development for AIBs is focussed on metal oxides and metal sulfides, the first attempt in developing copper hexacyanoferrate (CuHCF) as a cathode material for aqueous Al-ion battery was attempted by Liu et al. in 2014 [38]. Prussian blue analogs (PBAs) have been much explored as battery electrodes and as an electrochromic material [39] and CuHCF and nickel hexacyanoferrate (NiHCF) with a structure similar to PBAs also establish electrochemical properties [40]. In 2013, Reed et al., developed CuHCF cathode and tested its electrochemical performance in an organic aluminium trifluoromethanesulfonate (aluminium triflate) dissolved in diethylene glycol dimethyl ether (diglyme) as the electrolyte. An initial discharge capacity of 60 mAh g^{-1} was attained and gradual fading of the discharge capacity was noticed after 10 cycles [41]. PBAs is expected to have the high-rate capability, long cycle life and good electrochemical behavior [42], the synthesized CuHCF also has the face-centered cubic Prussian blue structure and expected to display a similar electrochemical nature. The efficiency of this newly developed system is claimed to carry more charge in AIBs because of the

trivalent Al ions compared to the other monovalent and divalent systems reported. The developed CuHCF electrode in aqueous aluminium sulfate reveals a discharge capacity of 63 mAh g⁻¹ at a current density of 50 mA g⁻¹. The specific capacity depends mainly on the Fe²⁺/Fe³⁺ redox couple in CuHCF electrode material [38]. The low discharge capacity using CuHCF as a cathode in the both aqueous and organic electrolyte may be owing to the cathodic and anodic passivation.

Cu_{0.31}Ti₂S₄ as cathode material with spinel structure delivered reversible capacity around 25 mAh g⁻¹ substantiating that TiS₂ can function as a better cathode material than Cu_{0.31}Ti₂S₄. Upon subsequent cycles the capacity is found to diminish from the first cycle, the reason being the rigid and less open crystal structure of cubic Cu_{0.31}Ti₂S₄ thwarting the Al intercalation [29]. A novel electrode material Cu_{2-x}Se nanorods were developed in an acidic ionic liquid electrolyte by Jiang et al. in 2018. The discharge capacity of 260 mAh g⁻¹ was obtained at a current density of 50 mA g⁻¹, upon further cycles, the capacity sustained approximately to 245 mAh g⁻¹ with 98% coulombic efficiency. It is speculated that the ultrahigh electronic conductivity of Cu_{2-x}Se nanorods (340 S cm⁻¹) assist in stimulating the capacity with high retention in coulombic efficiency [17, 44]. Added to this, the mechanism pertaining to the intercalation/de-intercalation of Al³⁺ ions during charging and discharging process was also validated. The redox reaction depends on the interconversion between Al₂Cl₇⁻ and AlCl₄⁻ ions, where Al₂Cl₇⁻ ions decompose to AlCl₄⁻ and Al³⁺ during the charging process and the reverse occurs during discharge. The structural change of Cu_{2-x}Se nanorods in each cycle with the intercalation and de-intercalation of AlCl₄⁻ ions is a quasi-reversible process [16].

A new kind of hybrid system by combining the chemistry of lithium (LiFePO₄) and aluminium in a single device was developed by and could be used for grid and stationary applications [44]. LiFePO₄ is one of the most widely used cathode materials till date in LIBs, owing to its stability, low cost, safety and durability [45]. Based on the works reported in LIBs with LiFePO₄ as cathode active material, hybrid battery with an aluminium anode, LiFePO₄ as a cathode and acidic ionic liquid (EMIMCl) was attempted which gives an initial capacity of 160 mA h g⁻¹. A special note to be mentioned in this hybrid system is there is no requirement of lithium salts since LiFePO₄ will preserve the required electrochemical reactions during the charging process for the successive cycles. The reversible capacity gradually decreases to 144 mA h g⁻¹ after 10 cycles which is further declined to 99 mA h g⁻¹ after 20 cycles. Lithium intercalation was confirmed as the foremost reaction occurring at the cathode in the course of the discharge process, except the observed voltage which is below the theoretical value challenging further insight into this [44]. Certain discrepancies were left for the present-day researchers, to have a better understanding of the reasons behind these and to develop electrode materials which could overcome these disputes.

Micro spherical CuS dispersed with acetylene black nanoparticles was synthesized by Wang et al, acetylene black will facilitate the charge transfer process. The integrated CuS@C based electrode was used as a cathode for aluminium ion batteries in AlCl₃/[EMIM]Cl ionic liquid electrolyte. The theoretical capacity of CuS@C as calculated from Faraday's law is 280.3 mAh g⁻¹ and practically, the capacity of about 240 mAh g⁻¹ was achieved [46]. On subsequent cycles, the discharge capacity declined which is due to the formation of solid electrolyte interface and irreversible capacity loss [47]. When the charge/discharge study was conducted for 20 consecutive cycles, the discharge capacity of 100 mAh g⁻¹ was acquired with similar discharge curves for all the cycles. Nearly 100% coulombic efficiency was achieved using integrated CuS@C as cathode material in AIBs which could deliver a specific capacity of 90 mAh g⁻¹ [46]. Another type of carbon coated cathode material for AIBs is reported by Jiang et al., Li₃VO₄ coated with carbon by the spray drying process. The initial discharge capacity of 137 mAh g⁻¹ attained at a current density of 20 mA g⁻¹, added to the development of a new material as cathode the authors have also targeted on figuring out the charge-discharge mechanism. Briefly, in the anode, the battery undergoes deposition and dissolution of metallic aluminium and insertion/de-insertion of Al³⁺ ions in the cathode. The amorphous V₂O₅/C composite was prepared by a zero-emission technique which reveal an initial discharge capacity of 150 mAh g⁻¹ when tested as a cathode material for AIBs. It is noteworthy to mention that the valence state of vanadium changes during the charging/discharging process [4]. Although the incorporation of carbon is expected to enhance the conductivity and in turn

the capacity of the electrode material, the results observed in this case is less compared to the other oxides of vanadium as discussed in table 1.

Graphene and graphene-based materials have attracted attention in the field of energy storage materials owing to their unique properties, making it a promising candidate for electrochemical energy devices [47, 48]. There are many reports available with the use of graphite as an electrode material for energy storage in ALBs and even offers high charge/discharge voltage [50], the role of graphite as an electrode material is not dealt in the present review as mentioned already. Based on the earlier studies on graphene/graphene-based materials, Ni₃S₂/graphene microflakes was developed and fabricated to study its electrochemical properties. The discharge capacity of 350 mAh g⁻¹ was achieved which declined down to 60 mAh g⁻¹ after 100 cycles sustaining a coulombic efficiency of 99% [51]. An alternative material for the current collector (tantalum) was also employed to overcome the poor electrochemical stability of metallic current collectors used so far [52]. Ni₃S₂/graphene electrode exhibits a stable capacity from 10th to 30th cycle which may be due to the stabilization of the other side reactions [53] and even at higher current density, the capacity lies between 235 to 300 mAh g⁻¹ [51]. Cobalt sulfide encapsulated with carbon nanotube (CNT) and carbon nanofiber film (CNF) (Co₉S₈@CNT-CNF) as a new electrode material was developed recently which reveal a high discharge capacity of 315 mAh g⁻¹. The composite with its porous structure enables more active sites leading to an enriched kinetic process in AIBs [54].

Hu et al., attempted a novel electrode material for AIBs by preparing freeze-dried reduced graphene oxide (rGO) supported SnS₂ as the cathode active material. Among the other hybrid materials developed so far, SnS₂/rGO offers the highest capacity, because of the larger surface area of SnS₂ and highly conductive rGO matrix which brings about improved ion-electron diffusion pathway. The initial discharge capacity of SnS₂/rGO was 392 mAh g⁻¹ which is high compared to bare SnS₂ with a capacity of 246 mAh g⁻¹. This clearly indicates that graphene in the cathode material plays an imperative role in facilitating the transportation of ions [55]. Figure 2 shows the comparison of initial discharge capacity with discharge capacity after cycles for the various hybrid systems. It is clear from the figure that hybrid systems developed using graphene, CNT depicts initial discharge capacity over 300 mAh g⁻¹ which has declined to 87 mAh g⁻¹ and 60 mAh g⁻¹ for CoS₂/CNT and Ni₃S₂/graphene respectively. In the case of Co₉S₈@CNT-CNF hybrid system even after 200 cycles the discharge capacity of 297 mAh g⁻¹ maintaining efficiency of 94% from the initial discharge capacity. It was reported that even after 6000 cycles the discharge capacity of 87 mAh g⁻¹ [54] was achieved, which is high compared to the discharge capacity of graphene/CNT based hybrid systems for 100 cycles.

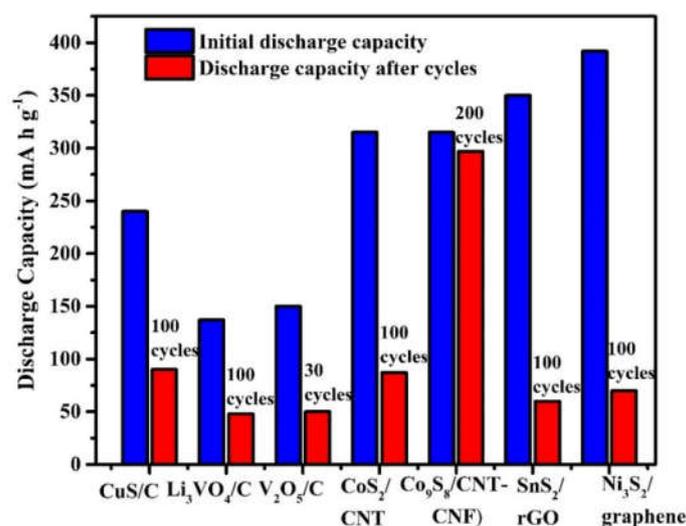


Figure 2. Comparison of discharge capacity initial and after cycles for various hybrid systems.

3. Future perspectives

In this mini-review, the cathode materials based on metal oxides, metal sulfides and hybrid systems based on the integration of metal oxides/metal sulfides with carbon-based materials (carbon, graphene, CNTs) and CuHCF used in AIBs have been summarized. It can be witnessed that the discharge capacity value reported in this review were diverse in all the cases, which establish that capacity depends on the cathode material engaged in each case. Based on all these studies it is suggested that researchers need to develop a cathode material with long cycle life, enhanced capacity, no side reactions, long stability, good coulombic efficiency and absence of structural disintegration during the intercalation and de-intercalation process. Novel graphene-based composite materials can overcome the mentioned aspects together with progress in the electrolyte and aid in the commercialization of AIBs which is still in the inception. Detailed analysis of the materials developed based on graphene its intercalation/de-intercalation mechanism, reaction kinetics, diffusion process should be addressed which should make up with the limitations of the already existing cathode materials.

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