



Aluminum-ion battery technology: a rising star or a devastating fall?

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As the share of renewable energy in the overall global energy consumption increases (e.g., the readers are kindly directed to the European Union goals [1–3]), issues of energy storage, alongside efficient electric transmission and distribution (using smart grid management [4]) are becoming the primary concerns. The electricity generated using renewable technologies (e.g., solar and wind energy sources) is being harvested upon catching the proper and available weather and daytime. Often, the moment of power generation does not meet the time this energy is needed for consumption [5]. Seeking cost-effective energy storage technologies is one of the main hindrances to the full implementation of renewable energy technologies as primary sources of energy. The energy storage technologies utilized nowadays can be divided into different categories, including chemical (such as hydrogen), electrochemical (various types of batteries), electrical (supercapacitors), mechanical (including compressed air and pumped hydro energy storage), and thermal (hot water storage and molten salt technologies) solutions [5]. Even though energy storage can be achieved in a variety of ways and methods, usually when considering small-scale energy storage systems for a short-term application, battery storage systems have the advantage of being easily distributed and simply modulated.

Additional to renewable energy storage, the increasing interest and demand for light-duty electric vehicles led to an enormous global research effort after new battery chemistries [6]. On the one hand, the well-known already commercialized lithium (Li)-ion battery (LiB) is increasing its global market share while demonstrating higher-energy densities with a significant cost drop to its lowest price per kWh every year [7]. However, when considering stationary energy storage facilities

and applications, the amount of energy that can be stored is directly proportional to the amount of active material. Therefore, for this kind of use, the cost per kWh is the determining factor, since massive quantities of raw material will be utilized [8]. Additionally, one should be aware that some significant challenges may result from the need to use aprotic organic solvents as constituents of LIB's electrolyte. Those electrolytes lead to several obstacles, including safety-related issues, capacity fading during cycling, and even issues in the production process due to the filling and wetting process required [9]. Moreover, the growing concern of the public and the companies in the matter of supply risk of mineral resources led the research community to investigate abundant materials as possible negative electrodes that are capable of providing increased safety levels, as well as higher theoretical energy densities [10].

One of the promising materials and relatively mature technology that has been in constant development and research in the last 20 years is magnesium (Mg)-based batteries [8, 11–14]. Besides being a multivalent anode, metallic Mg presents further significant characteristics. Low cost (its elemental abundance on the Earth's crust is significantly more than Li [13]), low negative potential, higher safety, and high theoretical volumetric and gravimetric storage capacities (3.83 Ah cm^{-3} and 2.2 Ah g^{-1} , respectively [8]) are only part of the advantages offered by the Mg anode. After the promising proof of concept achieved back in 2000, using Chevrel phases, many companies were interested in developing the technology and in the use of it for numerous applications [15]. On the other hand, various studies and reviews published lately have presented several barriers that seem to be challenging to overcome and hinder the commercialization of the Mg-ion battery technology [8, 14, 16].

The chemistry standing behind the use of a multivalent material, in opposition to Li and sodium (Na), induce certain issues related to electrolyte limitations and shortage in efficient cathode materials. Most of the challenges in the Mg-ion batteries arise from the high polarization ability of the divalent cations [8, 11, 13, 14, 17]. The higher charge density

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of Mg^{2+} ions, in comparison with Li^+ , leads to much stronger coulombic interactions with its surroundings, including the electrolyte and the active cathode material. Therefore, difficulties in reversible insertion and extraction of the divalent cation from the hosting material result in a sluggish migration and high-energy barriers. Financial efforts were also invested in plans that aim to overcome those difficulties. For example, E-MAGIC (FET-Open, European Magnesium Interactive Battery Community), a 4-year proactive project (with the Technion as one of the consortia members), was founded to demonstrate an innovative Mg-based energy storage technology.

Interestingly, even higher valent metal that has gained increasing attention in the last decade is aluminum (Al). Al seems like a promising technology as it is the most abundant metal on planet Earth and therefore presenting an affordable price along with high volumetric capacity in comparison with that of Li (8.05 in comparison with 2.04 Ah cm^{-3}), which are two primary considerations, particularly for portable applications [10, 11, 18]. Even more importantly, the extensive use of Al over the years for various applications has led to the development of a well-known and a cost-efficient recycling process, which is another major challenge existing for the Li-ion battery technology [19, 20]. But is it enough? With all the advantages listed (safety, recycling processes, and cost-effective prices) and holding the impressive feature of 4 times more energy per volume (compared with metallic Li), the development challenges associated with this technology are yet far from being resolved [21].

Although the number of research publications presenting innovative rechargeable Al-ion energy systems has increased dramatically in recent years, still plenty of uncertainty is present, and quite a large number of misconceptions are being delivered. Al-based systems can be separated into three main categories, including Al-air, Al-ion, chloroaluminate anion batteries [10], and a few additional systems such as Al-sulfur [22, 23] and Al-iodine [24, 25]. Primary Al-air batteries, which utilize Al metal against a porous carbon-based electrode that allows O_2 penetration, were studied using aqueous [26], non-aqueous [27], and ionic liquid-based electrolytes [28–31]. The pristine oxide layer of the Al metal alongside the parasitic reactions resulting from the direct interaction with the electrolyte media is a minor concern in comparison with the non-rechargeability obstacle that remains a challenge [18, 32].

Al-ion batteries can be described as batteries where Al^{3+} is the intercalating ion. This condition, alongside the facile deposition and dissolution of Al metal, is a key factor to reach the promising energy densities associated with the use of Al as the negative electrode [33]. Although various cathode materials were suggested and presented as possible hosts for Al^{3+} , still the discussions and demonstrated results dealing with

most of them lack the actual and correct mechanisms leading to the electrochemical activity exhibited (see Table 1). Mo_6S_8 , the Chevrel phase that was previously presented as a promising hosting material for Mg^{2+} ions, is one of the only compositions that were proven to be capable of hosting Al^{3+} ions both experimentally [34, 47] and theoretically, using bond valence energy landscape (BVEL) algorithm [21]. Recent calculations alongside experimental publications suggested a successful utilization of the $\alpha\text{-MoO}_3$ phase, as well [21, 35].

As presented in Table 1, most of the active cathode materials published allow reversible intercalation of chloroaluminate anions. Graphite and innovative carbon-based cathodes are the most studied and promising materials to serve in the Al-based energy storage systems. Theoretical calculations have shown no possible intercalation of the Al^{3+} ions, and the experimental publications struggle to prove it, as well [41–45, 51]. Several other known and less explored materials that were tested as possible cathodic materials presented an electrochemical performance that can be explained by either intercalation of solvated anions or later exposed side effects (see Table 1).

In summary, many efforts, both academic and financial (such as the ALION project [10]), are invested in the attractive Al-based energy storage technologies. The growing attention gave rise to an enormous amount of studies that claim to present promising innovative Al-ion hosting materials. Some of the research being published are of a poor scientific level, although being published in highly rated journals. Most of the publications reviewed here still fail to fully explain the electrochemical processes taking place throughout the cell's functioning. Moreover, a reasonable and adequate cycling capability, together with the promising theoretical storage capacities presented for the reversible Al^{3+} ion intercalation and metallic Al deposition processes, is yet far from being achieved. Clear differentiation between the various technologies, along with preliminary theoretical works, can hopefully lead to much focused and fruitful research. Experimental work involving a variety of characterization methods is mandatory to deeply understand and investigate the specific mechanism relevant to the electrochemical system.

It is our duty, as a responsible and matured research community, to be alert to the desire of some researchers to promptly publish results in high-impact journals, even in the cost of crossing a “red-line” of ethics and the essential need to verify the obtained results. We also must be on guard and take responsibility by accepting reviewing tasks and being alert for detecting flaws and incorrect data included in some of the manuscripts.

Table 1 A review of several reported cathode materials utilized for Al-based rechargeable systems

Cathode	Mo ₆ S ₈ Chevrel phase	MoS ₂	Fluorinated graphite	CuHCF	V ₂ O ₅	Graphite	NiS nanobelts
Electrolyte	1.5:1 AlCl ₃ - [BMIM]Cl	1.3:1 AlCl ₃ - [EMIM]Cl	0.5:1 AlCl ₃ - [BIM]Br	0.5 M Al(SO ₄) ₃ aqueous solution	1.1:1 AlCl ₃ - [EMIM]Cl	1.3:1 AlCl ₃ - [EMIM]Cl	1.3:1 AlCl ₃ - [EMIM]Cl
Discharge voltage	0.55 and 0.37 V	0.8 V	0.65 V	~ 0.6 V	0.55 V	1.8 V	0.9 V
Discharge current	12 mA g ⁻¹ (50 °C)	40 mA g ⁻¹	60 mA g ⁻¹	400 mA g ⁻¹	125 mA g ⁻¹	100 mA g ⁻¹	200 mA g ⁻¹
Initial capacity	148 mAh g ⁻¹	77.7 mAh g ⁻¹	~ 210 mAh g ⁻¹	41 mAh g ⁻¹	305 mAh g ⁻¹	~ 52 mAh g ⁻¹	104.7 mAh g ⁻¹
Reversible capacity	70 mAh g ⁻¹	66.7 mAh g ⁻¹	225 mAh g ⁻¹	22.5 mAh g ⁻¹ (the capacity gradually decreases)	273 mAh g ⁻¹	69.92 mAh g ⁻¹	~ 105 mAh g ⁻¹
Cycles	50	100	40	1000	20	100	100
Characterization method	Facile Al deposition/dissolution of Al was proved. CV of the carbon paper current collector with and without Mo ₆ S ₈ . CV presenting a two-step electrochemical reaction, indicating the two possible intercalation sites. Cracks in the active material were detected using SEM suggesting major expansion during cycling. ICP-OES and XRD evaluations provided further chemical and crystallographic evidence	XPS measurements indicate the presence of Al before and after cycling. Traces of Cl were also found to be present in the active material during cycling	A small shift in the XRD peaks when comparing the electrode after discharge to the pristine one—indicating enlargement of the space between the graphite sheets	XRD measurements have not shown any evident changes when comparing with the pristine structure electrochemical evidence is achieved	CV using two different electrolytes, proving that only when using AlCl ₃ :EMIMCl electrolyte, electrochemical evidence is achieved	CV of the Mo current collector with and without graphite. XPS and HRTEM evaluations before and after cycling	CV without the active material alongside XPS evaluations indicating the formation of Ni ₃ S ₂
Additional information	A follow-up work further investigates the two-step intercalation mechanism of the Al ³⁺ ions into the Chevrel phase [34]	Additional research, using MoS ₂ in aqueous electrolytes, have shown a significant dependency on the electrolyte's composition. Theoretical studies have supported the possible intercalation of Al ³⁺ in α-MoO ₃ [21, 35]	Previous studies using fluorinated graphite in non-aqueous Al-ion batteries failed to prove the Al ³⁺ intercalation into the graphite layers. No follow-up papers were found [36]	Additional studies performed using organic electrolytes have indicated the intercalation of the Al/diglyme complex into defect sites in the material [37]	The cathode was prepared using a stainless-steel current collector. The following studies have shown a battery-like performance that can be attributed to the iron and chromium present in the current collector [38]	DFT calculations indicate no possible intercalation of Al ³⁺ into the graphite layers [39, 40]. However, various studies demonstrated the intercalation of AlCl ₄ ⁻ into several carbon-based materials [41–45]	The Al ₂ S ₃ phase that is claimed to be formed during discharge was proven to be easily dissolved in chloroaluminate ionic liquids [46]
Year	2015	2018	2013	2015	2011	2015	2016
Ref.	[47]	[48]	[49]	[50]	[33]	[51]	[52]

CV cyclic voltammetry, XPS X-ray photoelectron spectroscopy, HRTEM high-resolution transmission electron microscopy, SEM scanning electron microscopy, XRD X-ray diffraction, ICP-OES inductively coupled plasma optical emission spectrometry

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