



## Advances and challenges in lithium-air batteries



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### HIGHLIGHTS

- The overall picture about the present lithium-air batteries is reviewed.
- The challenges of battery's electrolyte and electrodes are emphasized.
- Several possible research directions for performance improvements are highlighted.

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### ABSTRACT

Rechargeable lithium-air batteries have ultra-high theoretical capacities and energy densities, allowing them to be considered as one of the most promising power sources for next-generation electric vehicles. The technology has been honed in various ways over the years, but it still experiences critical issues that need to be addressed in order to make it commercially viable. For instance, its practical capacity, round-trip efficiency, and cycling life are among the factors that need to be improved. In this review, the developments of this type of battery are presented. In particular, the system levels of design that encompass the optimization of the battery's electrolyte and electrodes are discussed. More importantly, this report provides perspectives on achieving the desired battery performance to meet the demands of commercial viability.

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#### Abbreviations and acronyms

AAO	anodized aluminum oxide	ORR	oxygen reduction reaction
CNT	carbon nanotube	PC	propylene carbonate
CPL	composite protective layer	PVDF	polyvinylidene fluoride
DEGDME	diethylene glycol dimethyl ether	PVDF-HFP	poly(vinylidene fluoride-co-hexafluoropropene)
DEMS	differential electrochemical mass spectrometry	RH	relative humidity
DFT	density functional theory	RM	redox mediator
DMA	N,N-dimethylacetamide	RTIL	room temperature ionic liquid
DME	dimethoxyethane	SEI	solid-electrolyte interface
DMSO	dimethyl sulfoxide	SEM	scanning electron microscope
EC	ethylene carbonate	SPE	solid polymer electrolyte
ETPTA	ethoxylated trimethylolpropane triacrylate	TEGDME	tetraethylene glycol dimethyl ether
EV	electric vehicles	TEM	transmission electron microscope
FePc	iron phthalocyanine	TEMPO	2,2,6,6-tetramethylpiperidinyloxy
HMPA	hexamethylphosphoramide	TPFPB	tris(pentafluorophenyl) borane
LAGP	lithium aluminum germanium phosphate	TTF	tetrathiafulvalene
LATP	lithium aluminum titanium phosphate	VC	Vulcan carbon
LiTFSI	lithium bis(trifluoromethanesulfonyl)imide	XPS	X-ray photoelectron spectroscopy
OER	oxygen evolution reaction	XRD	X-ray diffraction

## 1. Introduction

Global warming and finite oil reserves are two issues that exert tremendous pressure on the traditional vehicle industry. Radically different from the conventional gasoline-fuelled vehicles, electric vehicles (EV), which are powered by rechargeable batteries, not only relieve the pressure on gasoline usage but perhaps more importantly, eradicate pollution generated from exhausts, lending hopes to the vehicle industry. To date, one of the most promising candidates for such power sources is the lithium-ion battery [1–3]. It has demonstrated a reasonable energy density (theoretical value:  $\sim 400 \text{ W h kg}^{-1}$ ), which is much higher than that of conventional lead-acid batteries ( $30\text{--}40 \text{ W h kg}^{-1}$ ) and nickel-cadmium batteries ( $40\text{--}60 \text{ W h kg}^{-1}$ ) [4]. As a result, lithium-ion batteries have been used as the power source for electric vehicles with achieving a driving distance of  $\sim 400 \text{ km}$  per charge [5]. Despite the progress accomplished to date, however, the specific energy densities of lithium-ion batteries are still far inferior to that of the conventional gasoline engines ( $\sim 13,000 \text{ W h kg}^{-1}$ ) [6]. Thus, the exploitation of new energy storage technologies for EVs is still a grand challenge.

In attempts to improve the energy density, much attention has been paid to metal-air batteries, especially lithium-air, aluminum-air, and zinc-air batteries [7]. In this type of batteries, oxygen is straight obtained from air, as opposed to storing an internal oxidizer, and the pure metal is used as the electrode instead of conventional intercalated materials. Theoretically, the energy density of metal-air batteries only relies on the metal electrode so that it can be significantly increased. Among the metal-air batteries, the lithium-air battery has garnered the most attention, predominantly because lithium is the lightest metal. This means that it has the highest theoretical capacity ( $3862 \text{ A h kg}^{-1}$ ), which corresponds to an energy density of  $11,680 \text{ W h kg}^{-1}$  for a potential of about 3.0 V. Even based on the consideration of the entire battery system, the energy density of  $\sim 1000 \text{ W h kg}^{-1}$  is still several times higher than that of lithium-ion batteries [8], showing the remarkable potential to completely replace gasoline in vehicles.

The fundamental chemistry of lithium-air batteries involves lithium dissolution and deposition on the lithium electrode (or anode) and oxygen reduction reaction (ORR) and oxygen evolution reaction (OER) on the air electrode (or cathode) [9]. Since the demonstration of a prototype rechargeable lithium-air battery by

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