

Environmental Impact Analysis of Aprotic Li–O₂ Batteries Based on Life Cycle Assessment

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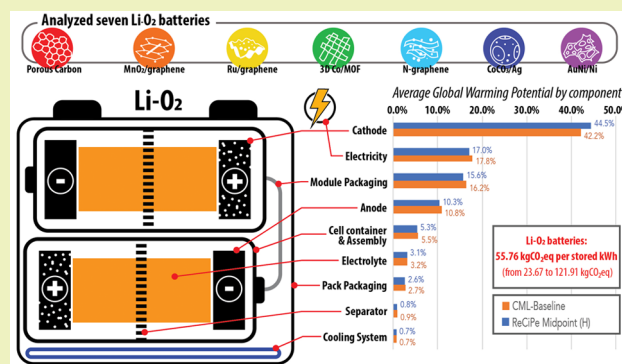
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ABSTRACT: Aprotic lithium–oxygen (Li–O₂) batteries are a prominent example of ultrahigh energy density batteries. Although Li–O₂ batteries hold a great potential for large-scale electrochemical energy storage and electric vehicles, their implementation is lagging due to the complex reactions occurring at the cathode. Great effort has been applied to find practical cathodes through the incorporation of different materials acting as catalysts. Here we tap into the quantification of the environmental footprint of seven high-performance Li–O₂ batteries. The batteries were standardized to feed a 60 kWh electric vehicle. Life cycle assessment (LCA) methodology is applied to determine and compare how different batteries and respective components contribute to environmental footprints, categorized in 18 groups. To get a bigger picture, results are compared with the environmental burdens of a reference lithium ion battery, reference sodium ion battery, and the average value of lithium–sulfur batteries. Overall, Li–O₂ batteries present lower environmental burdens in 9 impact categories, with similar impacts in 5 categories in comparison with lithium–sulfur and lithium ion batteries. With an average value of 55.76 kg-CO₂ equiv in Global Warming Potential for the whole Li–O₂ battery, the cathode is the major contributor, with a relative weight of 44.5%. These results provide a road map to enable the practical design of sustainable aprotic Li–O₂ batteries within a circular economy perspective.

KEYWORDS: Energy storage, Lithium–oxygen batteries, Life cycle assessment, Environmental impact, Ecodesign, Circular economy



INTRODUCTION

To meet the increasing energy demands, the development of efficient and clean energy storage technologies with a circular economy approach emerges as one of the foremost global challenges.^{1,2} In this context, electrochemical energy storage (EES) systems are one of the cornerstones for the development of a sustainable society based on renewable resources. Lithium-ion batteries (LIBs) hold the prevalent position and dominate the market of current EES because of their energy density and operation lifespan.³ However, current resource exhaustion, environmental pollution issues, and recent mandatory energy policies make desirable the development of EESs with ultrahigh energy density.^{4,5}

Since they were first reported in 1878,⁶ metal–air batteries emerge as a plausible solution, as notoriously large energy densities could be achieved thanks to concomitant metal oxidation and oxygen reduction.⁷ Rechargeable lithium–oxygen (Li–O₂) batteries containing nonaqueous aprotic electrolytes show a remarkable theoretical specific capacity of 3.860 mAh·g^{−1}, which represents a 5- to 10-fold increase regarding conventional LIBs.⁸ As a result, Li–O₂ batteries are among one of the most promising EES systems for large-scale electricity storage and hold a great potential to power electric

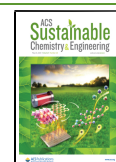
vehicles (EVs).⁹ Interestingly, EVs are resilient against petroleum supply chain disruption and can reduce the greenhouse gas emissions in comparison to conventional vehicles by up to 64%.¹⁰

Similar to Na-ion battery (NIB) and LIB half-cells,^{11,12} the configuration of Li–O₂ batteries consist of a metallic anode, a porous membrane acting as a separator soaked into a liquid electrolyte (it can also be gel-like or solid) and a porous cathode open to the ambient atmosphere.^{13,14} The basic difference between the well-established LIB or NIB technologies and Li–O₂ batteries is the cathodic process. Instead of the solid-state intercalation/deintercalation process, a catalytic reaction occurs in Li–O₂ batteries at the opened cathode.¹⁵ During such a process, Li⁺ ions react with the O₂ being supplied from the outside of the cell, forming Li₂O₂ as a cathodic product. During discharge, the oxygen reduction

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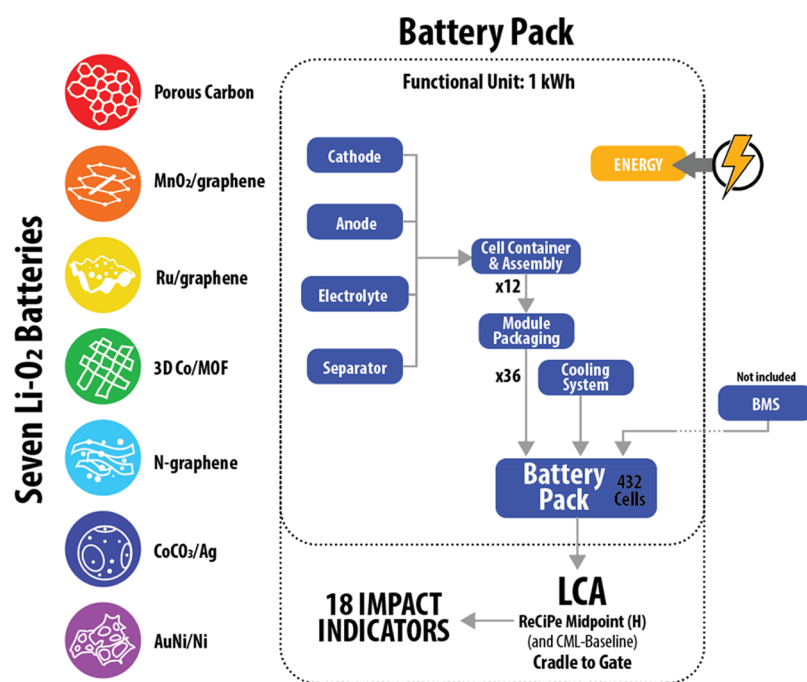


Figure 1. Battery component scheme and followed LCA scope and boundaries for analyzed seven Li–O₂ batteries.

reaction (ORR) occurs, while the oxygen evolution reaction (OER) take place during the charge phase.¹⁵

Unfortunately, the practical development of Li–O₂ batteries has been delayed because of the complex electrochemical reactions occurring at the cathode. At the early stage of research, Li–O₂ batteries have relied on the use of high surface area porous carbonaceous cathodes, which allowed large amounts of reactive sites for catalytic reactions.¹⁶ However, the Li₂O₂ formed during discharge can stop the electrochemical reactions at the same time that it enhances the overpotential required for its subsequent decomposition during charging.¹⁷ Additionally, a broad range of byproducts could be formed, and severe structural rearrangement at an atomic scale may occur upon charge/discharge. Therefore, over the past years, great attention has been focused to find practical cathodes through the incorporation of different materials which act as catalysts.

Li–O₂ battery development is still in the experimental stage, and there is still a long path left to realize their commercialization. Their commercialization is hampered by the safety issues related with the use of the highly reactive lithium metal anode, which shows the persistent dilemma of early Li dendrite formation can cause fire or explosion events. Such safety risk can be minimized implementing lithium alloy anodes (Li_xSi_y, for example) or using alternative gel or solid electrolytes.^{18,19} In this context, providing solid approaches toward the production of high-performance Li–O₂ batteries with reduced environmental impacts is of paramount relevance for the achievement of a sustainable electrified vehicle fleet.²⁰ The identification of environmental hot spots during Li–O₂ battery manufacturing would provide essential information toward battery redesign, developing energy storage systems with minimized energy consumption, lower environmental impacts and waste minimization.²¹ However, quantification of the environmental impacts of Li–O₂ batteries has been overlooked as most of the published manuscripts have been focused on the improvement of ORR and OER activities to

obtain decent reaction kinetics (to achieve large energy storage) with good reversibility (to afford long lifecycle). In spite of the low weight contribution of the cathode (nearly 2% when including electrolyte, anode, separator, current collectors, sealing and housing), its production can contribute up to 75% of the whole climate impact of a Li–O₂ battery,²² highlighting the urgent need to establish sustainable Li–O₂ cathode design approaches. More recently, Wang et al. have concluded that the environmental impacts of a Li–O₂ battery system having 63.5 kWh capacity are lower than those resulting from a reference LIB,²³ indicating the potential of Li–O₂ batteries for future EVs. These reports highlight the need for further research systematically quantifying the environmental impacts associated with the synthetic approaches required to obtain high-performance Li–O₂ batteries. As the most relevant methodology for the evaluation of environmental burdens of a product through its life cycle, life cycle assessment (LCA) can be applied to determine how different battery components contribute to environmental footprints.²⁴

Here we provide cues for future practical design of high-performance and sustainable rechargeable aprotic Li–O₂ batteries. To that end, the environmental impacts associated with the fabrication of seven state-of-the-art Li–O₂ batteries comprising different cathodes are analyzed in the framework of the LCA methodology. The environmental hot spots during Li–O₂ battery manufacturing are identified, allowing future improvement and stimulating the implementation of a circular battery economy.²⁵ The obtained results are compared with environmental impacts arising from other EESs, including mature LIBs and NIBs, as well as early-stage lithium–sulfur (Li–S) batteries. Although new configurations and chemistries could emerge in the near future due to the immature character of Li–O₂ batteries, we expect this study to be a guide to inspire further advances in the field of sustainable energy.

Table 1. Material and Energy Input Inventory for All the Studied Li–O₂ Batteries for a Standardized 60 kWh Battery Pack

mater.	Porous carbon		MnO ₂ /graphene		Ru/graphene		3D Co/MOF		N-graphene		CoCO ₃ /Ag		Au/Ni/Ni	
	w ^a (kg)	mater.	w ^a (kg)	mater.	w ^a (kg)	mater.	w ^a (kg)	mater.	w ^a (kg)	mater.	w ^a (kg)	mater.	w ^a (kg)	mater.
MSU ^b	2.88	MnO ₂ /GN ^b	2.25	Ru/PGE ^b	1.46	3D-MOF ^b	23.06	N-graphene ^b	3.55	CoCO ₃ /Ag ^b	3.55	all-metal ^b	1.15	
Al	2.65	GO ^c	0.26	graphite flakes	1.39	Co(NO ₃) ₂	23.06	GO	51.84	Co(NO ₃) ₂	5.84	Ni(CH ₃ COO) ₂ ·4H ₂ O	1.60	
Si	0.23	H ₂ SO ₄	0.03	NaNO ₃	1.39	H ₂ O	230.61	H ₂ O	86.39	H ₂ O	5539.07	hydrazine sulfate	0.82	
Al	2.65	KMnO ₄	2.25	H ₂ SO ₄	115.47	C ₄ H ₆ N ₂	123.13	melamine	155.50	K ₃ (Co(CN) ₆)	5.84	glycine	0.08	
FA ^d	6.54	H ₂ O	0.02	H ₂ O	1227.23	EO ^e	0.55			AgNO ₃	0.462	H ₂ O	21.61	
N	2.88	EtOH	0.01	H ₂ O ₂	12.13	PO ^f	1.27			EtOH	4320.47	chloroacetic acid	0.03	
NaOH	4.36	dH ₂ O ^g	0.02	EtOH	870.22	NaOH	0.2			Au			0.03	
EtOH	2.27			TEOS	15.73	RuCl ₃	0.04							
H ₂ O	6.53			Ac ₂ O	2.45									
C	1.44			DMM ^h	2.14									
PVDF ^f	0.48		0.26	HCl	1.5									
NMP ^j	7.20			EO	0.55									
				PO	1.27									
				NaOH	0.20									
				RuCl ₃	0.04									
				PTFE ⁱ	0.26									
Li ^k	7.04	Li ^k	7.04	Li ^k	7.04	Li ^k	7.04	Li ^k	7.04	Li ^k	7.04	Li ^k	7.04	
TEGDME	7.60	DMC	6.93	DMSO	5.18	DMSO	4.19	DMSO	47.95	TEGDME	3.94	TEGDME	65.66	
LiClO ₄	1.90	EC	8.55	LiClO ₄	0.07	LiClO ₄	0.02	LiClO ₄	7.47	LiPF ₆	1.13	LiPF ₆	18.58	
CaCl ₂	7.60	LiPF ₆	0.98					CHF ₃	1.13	CHF ₃	1.13	CHF ₃	18.58	
GF ^l	17.57	PP	8.87	GF	8.93	GF	7.03	PP/C2400 ^m	5.25	GF	7.22	GF	38.95	
Al ⁿ	20.74	Al ⁿ	27.01	Al ⁿ	10.54	Al ⁿ	8.30	Al ⁿ	15.98	Al ⁿ	8.52	Al ⁿ	45.98	
PP	6.33	PP	8.24	PP	3.22	PP	2.53	PP	4.88	PP	2.60	PP	14.03	
PE	4.12	PE	5.36	PE	2.09	PE	1.65	PE	3.17	PE	1.69	PE	9.13	
Ar	1.76	Ar	1.61	Ar	0.84	Ar	1.08	Ar	2.03	Ar	1.00	Ar	4.33	
metal F. (units)	7.76 × 10 ⁻⁸	metal F. (units)	7.10 × 10 ⁻⁸	metal F. (units)	3.68 × 10 ⁻⁸	metal F. (units)	4.74 × 10 ⁻⁸	metal F. (units)	8.93 × 10 ⁻⁸	metal F. (units)	4.39 × 10 ⁻⁸	metal F. (units)	1.90 × 10 ⁻⁷	
chem. F (units)	5.71 × 10 ⁻⁸	chem. F (units)	5.22 × 10 ⁻⁸	chem. F (units)	2.71 × 10 ⁻⁸	chem. F (units)	3.49 × 10 ⁻⁸	chem. F (units)	6.57 × 10 ⁻⁸	chem. F (units)	3.23 × 10 ⁻⁸	chem. F (units)	1.40 × 10 ⁻⁷	
Al ⁿ	2.89	Al ⁿ	2.83	Al ⁿ	1.45	Al ⁿ	1.87	Al ⁿ	3.57	Al ⁿ	1.73	Al ⁿ	7.51	
Cu ^o	0.59	Cu ^o	0.58	Cu ^o	0.30	Cu ^o	0.38	Cu ^o	0.73	Cu ^o	0.35	Cu ^o	1.53	
IC	0.28	IC	0.28	IC	0.14	IC	0.18	IC	0.35	IC	0.17	IC	0.74	
ABS	1.26	ABS	1.24	ABS	0.63	ABS	0.82	ABS	1.56	ABS	0.76	ABS	3.28	

Table 1. continued

Porous carbon		MnO ₂ /graphene		Ru/graphene		3D Co/MOF		N-graphene		CoCO ₃ /Ag		Au/Ni/Ni	
mater.	w ^a (kg)	mater.	w (kg)	mater.	w (kg)	mater.	w (kg)	mater.	w (kg)	mater.	w (kg)	mater.	w (kg)
PP	0.65	PP	0.64	PP	0.33	PP	0.42	PP	0.81	PP	0.39	PP	1.70
IC ^p	0.62	IC	0.61	IC	0.31	BMS (Not Included in the LCA)	0.40	IC	0.77	IC	0.37	IC	1.61
Al ^r	0.21	Al ^r	0.20	Al ^r	0.10	Al ^r	0.13	Al ^r	0.26	Al ^r	0.12	Al ^r	0.54
Al ^r	1.61	Al ^r	1.58	Al ^r	0.81	Cooling System	1.04	Al ^r	1.99	Al ^r	0.97	Al ^r	4.19
EG ^q	4.16	EG	4.06	EG	2.09	EG	2.69	EG	5.13	EG	2.49	EG	10.79
Al ^r	8.81	Al ^r	8.60	Al ^r	4.42	Pack Packaging	5.69	Al ^r	10.86	Al ^r	5.27	Al ^r	22.85
steel ^r	0.52	steel ^r	0.51	steel ^r	0.26	steel ^r	0.34	steel ^r	0.65	steel ^r	0.31	steel ^r	1.36
Cu ^o	0.19	Cu ^o	0.18	Cu ^o	0.09	Cu ^o	0.12	Cu ^o	0.23	Cu ^o	0.11	Cu ^o	0.48
1337.37		1286.16		788.12		869.98		829.80		777.89		3468.68	
104.82		95.82		49.74		67.90		120.61		59.31		257.08	

^aw denotes weight. ^bActive material. ^cGO = graphene oxide. ^dFurfuryl alcohol. ^eEO = ethylene oxide. ^fPO = propylene oxide. ^gdH₂O = deionized water. ^hDMM = dimethoxymethane. ⁱBinder. ^jNMP = N-methyl-2-pyrrolidone (solvent). ^kLi billet, primary. ^lGF = glass fiber. ^mPP/C2400 = polypropylene/Celgard 2400 membrane. ⁿAl ingot. ^oCu wire. ^pIC = integrated circuit. ^qEG = ethylene glycol. ^rSteel, billet.

■ EXPERIMENTAL SECTION

Goal, Scope, Battery Selection, and Inventory Analysis.

Here, the environmental impacts of seven different rechargeable Li–O₂ batteries containing aprotic (nonaqueous) electrolytes are analyzed, quantified, and compared. As detailed below, such batteries were selected because they successfully improve the ORR and OER activities of Li–O₂ batteries following different strategies, which is essential toward commercialization.²⁶

Environmental impacts are categorized into 18 impact categories (called midpoint indicators) within the ReCiPe 2016 method. Such 18 midpoint (hierarchical) indicators were selected because they use impact mechanisms that have global scope. Unlike other approaches such as Eco-Indicator 99, the ReCiPe 2016 Midpoint method assumes that the impacts from future extractions have been included in the inventory analysis.²⁷ Midpoint indicators present a low uncertainty level when comparing with end point indicators while offering a mean for comparing the different impact categories typically used in LCA. More precisely, global warming potential (GWP) [kg·CO₂ equiv] has been used as a mean to obtain a cross-field comparison with other EES systems.²⁰ The scope and boundaries of this study are schematically shown in Figure 1.

Table 1 provides a summary for the material and energy input inventory of analyzed Li–O₂ batteries (such information is extracted from laboratory data). An additional disclosure of the inventory modeling of battery materials production is provided in the Supporting Information (Tables S1 to S14). These batteries have been selected according to their cathode, which plays a pivotal role in the electrochemical performance. It should be noted that the configuration of aprotic Li–O₂ batteries substantially differs from that of commercial LIBs. To enable the O₂ reduction to solid Li₂O₂ on discharge and the reverse oxidation on recharge, an opened cathode is required in contrast with the solid and thick LIB cathodes. Figure 2 schematically depicts the components and electrochemical

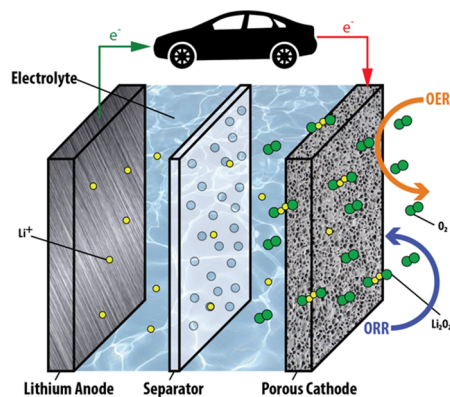


Figure 2. Schematic representation of the components and electrochemical processes in an aprotic Li–O₂ battery with a Li metal anode, a porous air cathode, and a separator soaked in an aprotic electrolyte. Upon discharge, Li⁺ ions move from the anode to the cathode side, releasing electrons to the external circuit and powering an electric vehicle.

processes in a Li–O₂ battery. Typically, aprotic Li–O₂ batteries present a lithium metal foil as the anode (negative electrode), a porous cathode (positive electrode), and an electrolyte in between formed by lithium salts dissolved in a nonaqueous solvent.²⁸ The cathode is usually an electronically conducting porous matrix that enables the electrochemical contact between O₂ and Li⁺ in the electrolyte. Li–O₂ functions thanks to the dissolution/deposition of Li metal at the anode and ORR/OER reactions at the porous cathode.²⁹ The specific electrochemical pathways, described by Abraham et al.,^{13,14} involve the reduction of molecular oxygen to lithium superoxide (LiO₂) via one-electron transfer as



After reduction, either disproportionation (eq 2) or receipt of a second electron (eq 3) can occur, yielding lithium peroxide (Li₂O₂) as the main discharge product:



Unfortunately, the limited solubility of Li₂O₂ in aprotic solvents can lead to premature cell death as a result of passivation and cathode clogging effects (blocking of the oxygen pathway) upon discharge. Upon charge, the deposited Li₂O₂ undergoes oxidation at the porous cathode surface, regenerating molecular oxygen. However, the sluggish oxidation of insulating Li₂O₂ at the positive electrode makes necessary high overpotentials, yielding detrimental side reaction products such as LiOH and Li₂CO₃.³⁰

Following this common design, numerous different cathode designs have been pursued to obtain high-energy-density Li–O₂ batteries. Thanks to their high electrical conductivity, large specific surface area, and porous architecture, early attempts on Li–O₂ cathodes focused their effort toward large surface area carbon nanomaterials. Among the different conductive carbon materials reported, mesoporous carbon mixed with Super P carbon showed an interesting specific capacity of ~9000 mAh·g⁻¹ when cycled at 0.005 C (battery 1, denoted as “porous carbon”).³¹ In spite of obtained performance, the cathode could only withstand 65 cycles.

Pure carbonaceous cathodes show low cycling stability and premature battery death due to their low activity toward Li₂O₂ decomposition, poor stability at high potentials, formation of insulating Li₂CO₃, and other side reactions leading to cathode surface passivation.³² Hybridization of carbonaceous structures with catalysts to enhance oxygen reduction/evolution reaction efficiency while maintaining high conductivity and chemical/electrochemical stability has been sought. Because of their intrinsically good OER and ORR activity, abundance, and low toxicity, manganese (Mn)-based oxides are the most investigated transition metal oxides in this sense.³³ Among reported Mn oxide/carbon nanocomposites, α-MnO₂/graphene shows especially remarkable characteristics for Li–O₂ batteries. α-MnO₂ nanorods were prepared through an *in situ* oxidation–reduction method onto graphene to obtain both improved electronic conductivities and increased active sites (battery 2, denoted as “MnO₂/graphene”).³⁴ As a result, a battery capable of delivering a specific capacity of 11 520 mAh·g⁻¹_{carbon} at 200 mA·g⁻¹ (0.06 mA·cm⁻²; 1 C = 10 A·g⁻¹) was achieved. Although the cycle life needs to be further improved toward practical application, the battery provided 2900 mAh·g⁻¹ after 25 cycles at 300 mA·g⁻¹. Noble metals, known for their excellent catalytic properties, have also been used for Li–O₂ battery cathodes. For example, ruthenium nanocrystals were incorporated into porous graphene to promote oxide evolution reactions, providing a reversible capacity of 17 710 mAh·g⁻¹ with long cycle life (1000 mAh·g⁻¹ after 2000 cycles) (battery 3, denoted as “Ru/graphene”).³⁵

Additive manufacturing technologies allow the fabrication of hierarchical materials with tailored geometries. These technologies also exhibit lower energy consumption and comprise shorter manufacturing processes, reducing the environmental impacts during fabrication.³⁶ Using an extrusion-based 3D printing technique, a novel approach for the fabrication of porous carbon structures with embedded Co nanoparticles as catalysts was reported (battery 4, denoted as “3D Co/MOF”).³⁷ Upon carbonization of the 3D-printed metal–organic framework with Co nanoparticles, the porous cathode provided enough pore space for Li₂O₂ storage and intimate contact between Li₂O₂ and the electrocatalysts, delivering 1124 mAh·g⁻¹_{electrode} at 0.05 mA·cm⁻² and a specific energy of 798 Wh·kg⁻¹, in comparison with the <300 Wh·kg⁻¹ characteristic of LIBs.

Heteroatom doping is a facile and efficient approach to improve the catalytic activity of carbon by modifying its surface/electronic properties. This could be done through simple and cost-effective hydrothermal and subsequent annealing processes. In this context, a

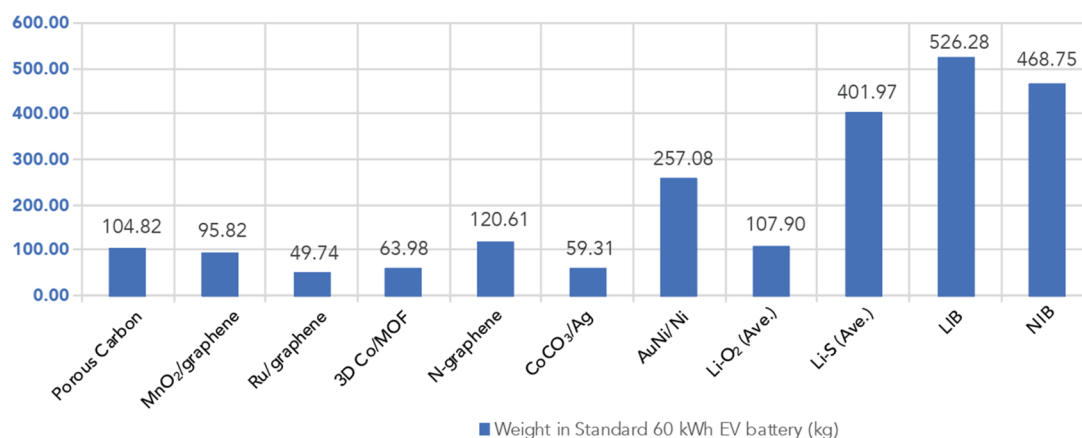


Figure 3. Standardization of weights for studied Li–O₂ batteries for a 60 kWh EV battery. LIB data are obtained from Ecoinvent 3.7 database, while NIB and Li–S data are extracted from refs 44 and 24, respectively.

hierarchical porous nitrogen-doped 3D graphene cathode with increased active sites and large surface area for oxygen diffusion and electrolyte infiltration was prepared (battery 5, denoted as “N-graphene”).³⁸ A specific capacity of 7300 mAh·g⁻¹ at 50 mA·g⁻¹ was achieved in comparison with the 2250 mAh·g⁻¹ capacity of the non-doped graphene. Moreover, a stable cycling performance was obtained during 21 cycles in comparison with the 8 cycles achieved for the bare graphene.

Side reactions with Li₂O₂ have been avoided upon the fabrication of carbon-free cathodes. Accordingly, hybrids based on Co₃O₄@Co₃O₄/Ag having a yolk–shell structure were fabricated via a synchronous reduction process to obtain active sites with higher ORR and OER activity (battery 6, denoted as “Co₃O₄/Ag”).³⁹ As a result of the favorable Li₂O₂ diffusion and decomposition, the electrical conductivity of Ag, and the porous character of Co₃O₄, an outstanding initial capacity of 12 000 mAh·g⁻¹ when cycled at 200 mA·g⁻¹ was achieved. Additionally, a long cycle life (1000 mAh·g⁻¹ after 80 cycles at 200 mA·g⁻¹) was achieved because of the synergetic effects arising from interfacial/surface interactions and doping engineering.

Not only high specific capacities but also long life cycles are required to enable energy storage systems with lower environmental impact.⁴⁰ This is especially relevant in Li–O₂ batteries, where resolving the cathode corrosion by formed Li₂O₂ and LiO₂ is a serious bottleneck for practical implementation. To that end, Xu et al. hybridized an alloy comprising gold and nickel (AuNi) with nanoporous Ni and further supported onto a Ni foam to obtain an electrically conducting porous structure where AuNi acted as an ORR and OER catalyst (battery 7, denoted as “AuNi/Ni”).⁴¹ As a result, an outstanding capacity of 22 551 mAh·g⁻¹_{AuNi} (the highest reported to date) was achieved. Interestingly, a remarkable life cycle was achieved, with 3000 mAh·g⁻¹_{AuNi} after 286 cycles at 1 A·g⁻¹.

For battery manufacturing, our model includes the use of a glovebox containing argon gas to provide a moisture-free inert atmosphere during battery cell assembly (argon consumption proportionally assigned with a reference of 600 kg for a battery production of 162 kg·day⁻¹), which is a predominant driver of energy use during battery manufacturing.⁴² Additionally, the impacts associated with both metal and chemical factories are also taken into account during cell assembly (“item” as the unit). The electricity consumed during production has been computed as 17.20 kWh·kg⁻¹ based on the analysis reported by Deng et al. because it provides an estimate value of the future large-scale industrial production of postlithium batteries for electric vehicles.⁴³ The process includes mixing, coating and drying, calendaring, notching, stacking, welding, and sealing, electrolyte injection, final cutting/degassing/sealing/pouch forming, and a dry room (4000 m²). The electricity required for each material within the inventory is included within each component in the Ecoinvent 3.7 database.

Regarding the remaining battery components, the anode is composed of pure metallic lithium foil, which in the case of the CoCO₃/Ag battery has been blended together with carbon as the conducting phase and poly(vinylidene fluoride) (PVDF) as a binder. As PVDF separators commonly used in LIBs can react with Li₂O₂ to form unwanted discharge products, these batteries use either a glass microfiber or polypropylene separator, usually known as Whatman and Celgard (respectively) in the battery field. Such separators are soaked into a nonaqueous liquid electrolyte (tetraethylene glycol dimethyl ether (TEGDME), dimethyl carbonate (DMC), ethylene carbonate (EC), or dimethyl sulfoxide (DMSO)) having a variety of dissolved lithium salts (CF₃SO₃Li, LiPF₆, LiClO₄, or lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) depending on the battery choice). Cell container and assembly, module pack packaging, cooling system, and packing are composed of the same materials, and their quantities differ in terms of battery sizes. A battery management system (BMS) has not been included in the study since it is electrochemically inactive; it may obstruct a proper quantification of the environmental impacts of Li–O₂ batteries. Likewise, inventory modeling does not take into account the implementation of gas inlet and outlet valves on the cathode side to afford the transport of oxygen. We estimate that the fact of not taking these common elements into account (to the seven studied batteries) allows us to obtain greater accuracy when comparing the results.

Life Cycle Interpretation. LCA analyses were done with OpenLCA software using Ecoinvent 3.7 Data set.¹⁰ A cradle-to-gate perspective was followed as a result of the nonmature character of Li–O₂ batteries and the current lack of data regarding their recycling.⁴⁴ A cradle-to-grave analysis allows detection of ~82% of the kg·CO₂ equiv emitted by the battery during its life, and 62–99% in the rest of the 11 impact categories.²⁴ Although the material and energy input inventory have been calculated for a 60 kWh battery pack (typically required to power an electric vehicle), 1 kWh of storage capacity was used as a functional unit (FU) to facilitate any comparison with other EESs (including sodium ion⁴⁴ and lithium–sulfur batteries).²⁴

Comparison with Lithium Ion and Lithium–Sulfur Batteries. To determine whether or not Li–O₂ battery technology is more environmentally sustainable than other relevant battery systems, the obtained environmental impacts were compared with the results reported for a lithium manganese oxide (LiMn₂O₄, LMO) LIB, NIBs, and Li–S batteries. An LMO-based LIB was selected because it is a well-established chemistry that offers a good cost balance in comparison with that of other emerging LIB chemistries, providing a solid base for comparison. Moreover, this cathode offers a high capacity, a relatively low toxicity, and use of naturally abundant Mn. NIBs were taken into account due to their potential to replace LIB for stationary applications, and Li–S technology was chosen as a representative ultra-high-energy-density EES based on lithium. Data for LIB are extracted from the reference compact electric passenger car (918.22 kg with no battery in Ecoinvent 3.7 database).¹⁰ The

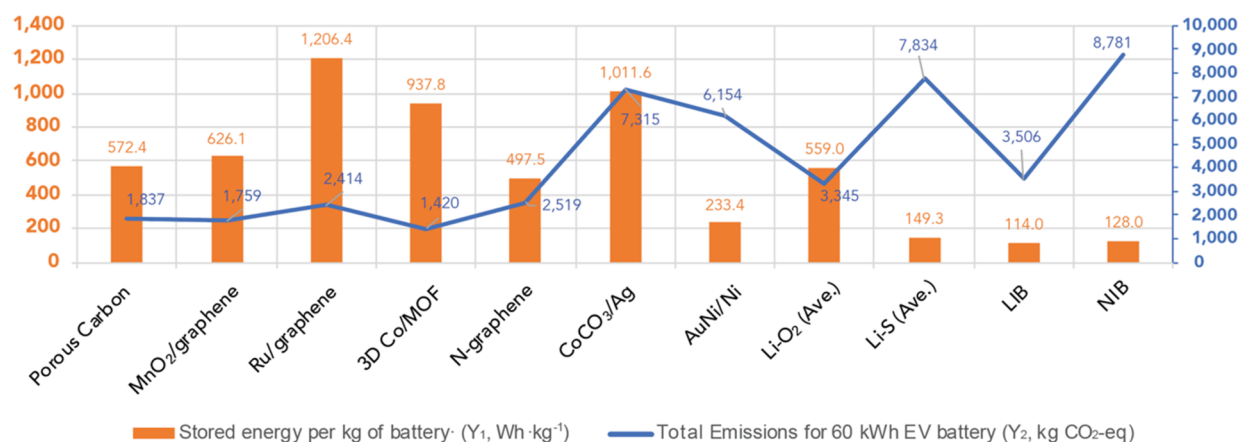


Figure 4. Storage capacity in 1 kg of battery and total kg-CO₂ equiv emissions for a standardized 60 kWh EV battery. LIB data are obtained from Ecoinvent 3.7 database, while NIB and Li-S data are extracted from refs 44 and 24, respectively.

battery is a prismatic LIB having a LMO cathode together with a polyethylene separator, graphite-based anode, and LiPF₆ in ethylene carbonate electrolyte. With 262 kg, the battery pack contains 14 single cells and provides an electric supply of 29.87 kWh with 48 V (denoted as the “LIB reference case” from now on). The environmental impacts for NIBs were obtained from the work done by Peters et al., who analyzed the LCA of a common NIB system battery consisting in a layered oxide cathode in combination with a hard carbon anode (1 kWh of storage capacity as a functional unit; NaPF₆ in an organic solvent electrolyte; polyethylene/polypropylene porous sheet separator).⁴⁴ Data for Li-S batteries were extracted from a recent research work by our group where the environmental impacts of five diverse lithium-sulfur batteries have been studied in a cradle-to-gate LCA.²⁴ Li-S batteries deliver a power of 57–62 kWh and contain a metallic Li anode, a polypropylene separator, an electrolyte based on LiTFSI, dioxolane, dimethoxyethane, and lithium nitrate, and an aluminum/polyethylene/polypropylene cell container.

RESULTS AND DISCUSSION

Li-O₂ Battery Weight Estimation. For a given energy storage capacity, Li-O₂ batteries have a remarkably low weight in comparison with those of LIBs. As vehicle weight reduction is among the most efficient routes to achieve large cumulative emissions,⁴⁵ we first evaluate the total weights of the seven Li-O₂ batteries standardized for a 60 kWh battery pack. Figure 3 shows the weight standardization of Li-O₂ batteries for a 60 kWh EV battery (data extracted from Table 1). For the sake of comparison, weights corresponding to a 60 kWh battery based on Li-S,²⁴ LIB (Ecoinvent 3.7 database), and NIB,⁴⁴ are also shown. LIB results a 12% heavier than a 60 kWh EV comprising a NIB due to the relatively poor energy density (114 Wh·kg⁻¹) of the LMO cathode, which remains below the 128 Wh·kg⁻¹ delivered by the NIB with a layered oxide cathode in combination with a hard carbon anode.⁴⁴ Li-O₂ batteries are up to 11 times lighter than LIBs. More precisely, a 60 kWh LIB pack weighing 526.3 kg could be potentially replaced by a 50–260 kg Li-O₂ battery pack, constituting a great benefit considering that the gross weight of an automobile is typically within the 1500–2500 kg range.⁴⁶ Among the seven batteries studied herein, Au Ni/Ni is the heavier battery pack due to used cathode materials. On the contrary, cathodes based on graphene allow lightweight Li-O₂ batteries thanks to the good electrochemical performance arising from the high electrolyte contact area, electron transport rate and structural stability provided by graphene.⁴⁷

Figure 4 shows the storage energy per 1 kg of battery together with the total kg-CO₂ equiv emissions for each of the studied batteries. As a general trend, the batteries having the lowest gravimetric energy densities require larger amount of material to fulfill their requirements, where Li-S, LIB, NIB, and Au Ni/Ni are the heaviest battery packs. Generally, the kg-CO₂ equiv total emissions for each battery increases as both the weight of the battery increases and the storage potential decreases. The total emissions for a 60 kWh LIB battery pack are nearly 3506 kg-CO₂ equiv, while reduced values of 1400–2500 kg-CO₂ equiv are achieved for most of the analyzed Li-O₂ batteries. Notably different CO₂ equiv is noted among analyzed Li-O₂ batteries, where CoCO₃/Ag and AuNi/Ni have the highest impact. Variations among same battery technologies were already detected in Li-S batteries, where despite the high average GWP value of 7834 kg-CO₂ equiv for a 60 kWh battery, NaCMC-Sulfur has a reduced impact in the range of ~3200 kg-CO₂ equiv, which is below than that corresponding to a LMO-based LIB.²⁴ Moreover, the EV weight savings promoted by Li-O₂ battery weight reduction provide an immediate reduction on the environmental impacts during the operational phase,⁴⁸ providing a reliable approach to lower the environmental impacts of batteries for EVs independently of the electricity grid character. However, to obtain definite and decisive conclusions, further and more detailed analysis is required for a better understanding of the environmental impacts of each battery pack, not only during use but also taking into account material synthesis and battery pack manufacturing.

LCA for Li-O₂ Batteries. On the basis of the data shown in Table 1 and Figure 1, the cradle-to-gate environmental impacts of seven state-of-the-art Li-O₂ batteries per 1 kWh of storage capacity are shown in Figure 5. Impacts are categorized into 18 indicators (horizontally arranged) obtained from the life cycle impact assessment (LCIA). Depending on the impact category, increases up to a factor of ~60 are observed. These results reflect the fact that the different synthetic approaches markedly transfer and even reduce the environmental impacts of resulting Li-O₂ batteries.

3D Co/MOF battery shows the lowest scores in 15 of the impact categories (including GWP, one of the most representative midpoint impact categories). Such an outcome may arise from the fact that in this design the catalyst Co nanoparticles are embedded into a cobalt-based metal-organic

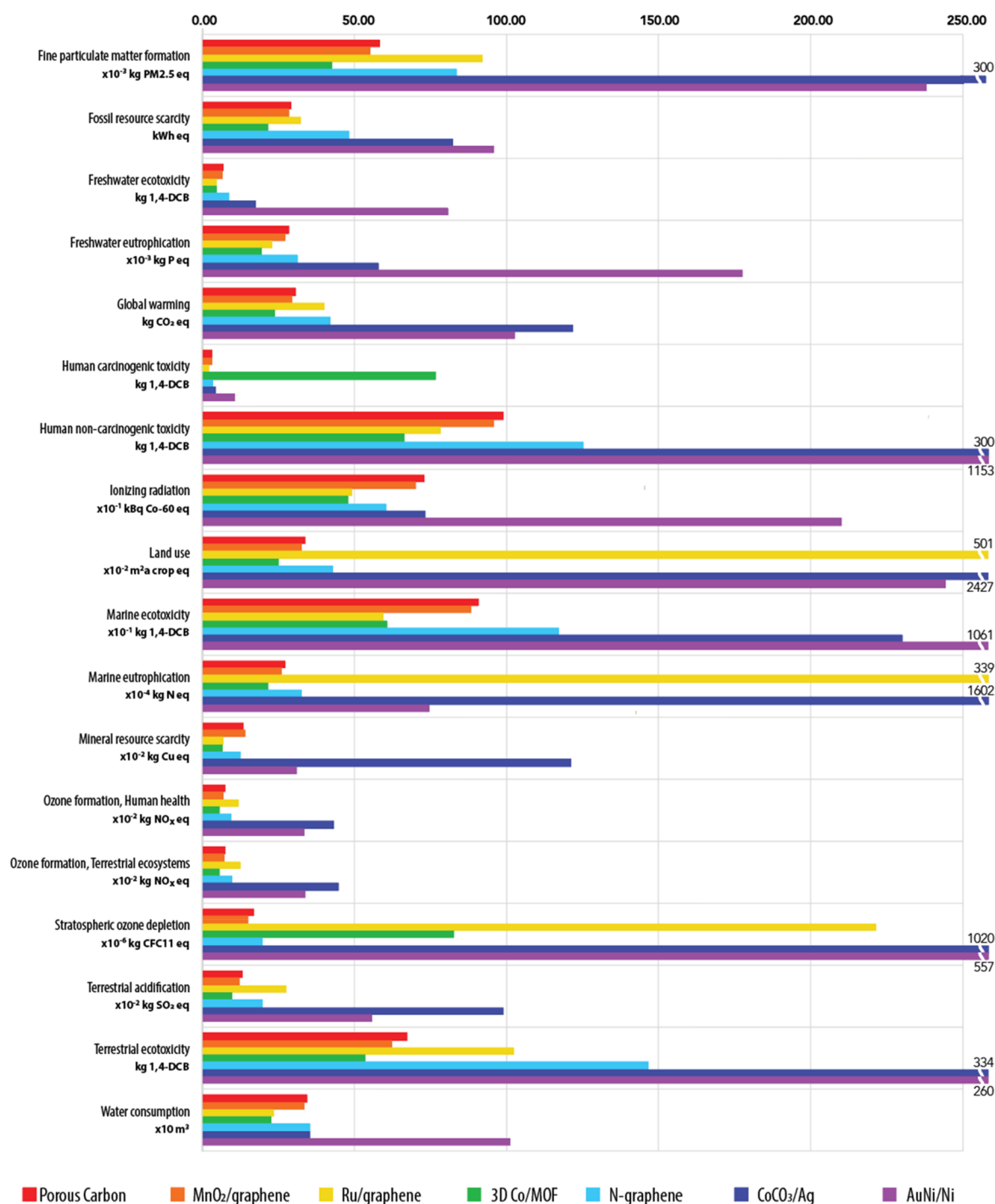


Figure 5. Environmental impacts of the Li–O₂ batteries per 1 kWh of storage capacity.

framework using an extrusion-based printer. This extrusion-based process requires low amount of energy for cathode fabrication, yielding decreased impact values. Due to their simplicity, process efficiency and use of rather common materials, porous carbon and MnO₂/graphene batteries are the next lower in environmental impacts. Ru/graphene offers large impacts in land use, marine eutrophication, stratospheric ozone depletion, and terrestrial ecotoxicity. Ruthenium is a rare transition metal (found in ~100 parts per trillion) and its compounds are considered highly toxic.⁴⁹ Overall, the battery systems showing larger environmental burdens are CoCO₃/Ag and AuNi/Ni, dominating impacts in 16 of the 18 categories

analyzed. Such results can be explained by the toxicity of employed materials, where the potential supply chain bottleneck associated with critical raw materials (CRMs) such as cobalt and nickel increases the environmental impact of the whole Li–O₂ battery.⁵⁰ Additionally, CoCO₃ is harmful to human health (irritating eyes and skin), while nickel can produce human death.⁵¹ In comparison with the other analyzed batteries, AuNi/Ni requires a large amount of energy for its fabrication and uses a substantial amount of aluminum (for the cooling system and cell-module-pack packaging). Finally, the N-graphene battery obtained through hydrothermal/annealing processes presents intermediate environ-

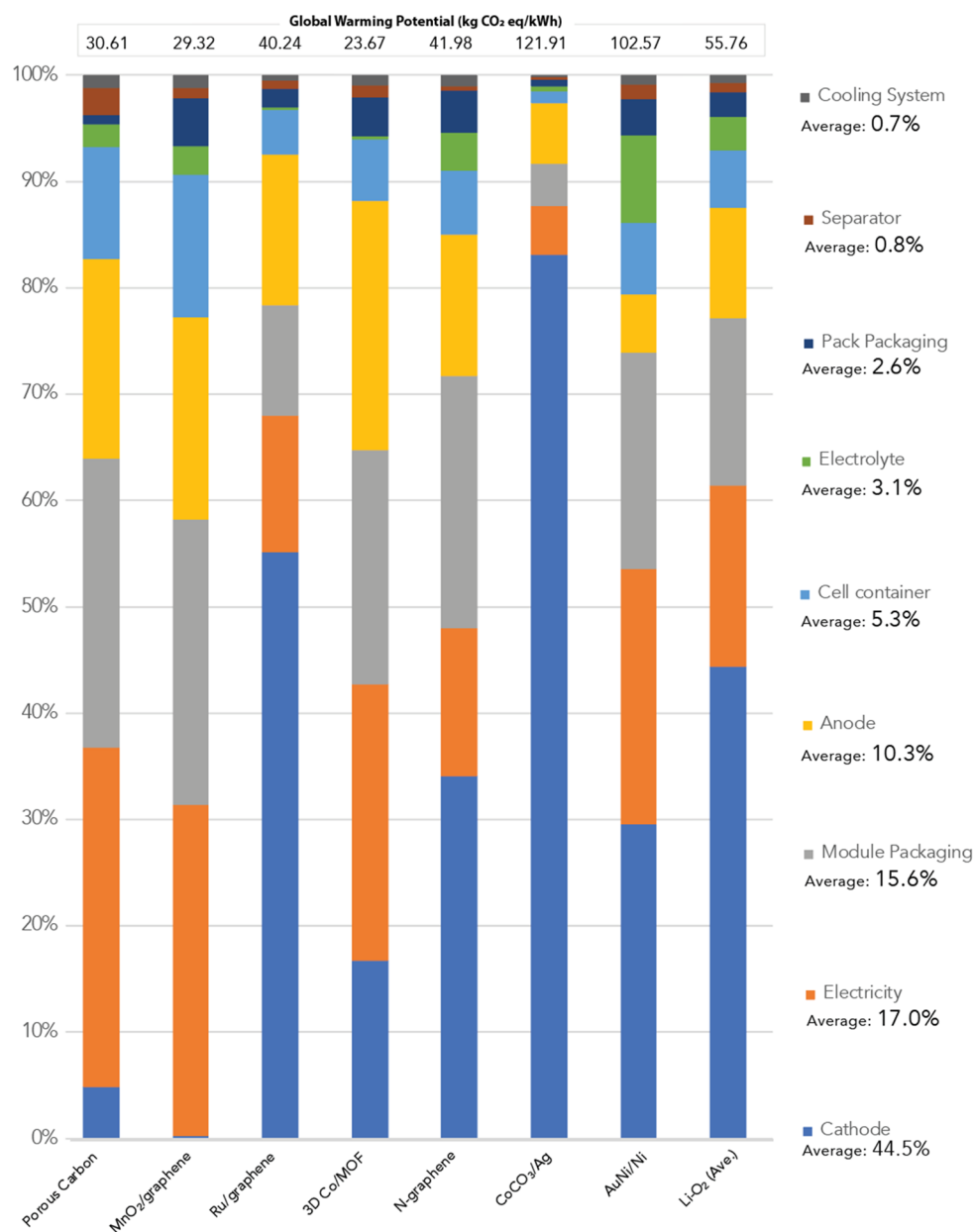


Figure 6. Relative contribution of each battery component to the global warming potential (GWP).

mental impact. Although graphene could exert a considerable toxicity regarding the environmental and human health, it provides a means to substitute scarce metals by nanomaterials based on the abundant element carbon.⁵²

As aprotic solvents (TEGDME, DMC, EC, or DMSO) are flammable and represent notable issues toward both environmental and human health, as a general trend, batteries presenting lower amounts of electrolyte present reduced impacts. In this sense, the AuNi/Ni battery requires almost 9 times more TEGDME than does the porous carbon battery. AuNi/Ni also needs large quantities of LiTFSI, whose synthesis requires large amounts of methyl chloride solvent, encompassing significant ozone depletion potential.⁴³ In spite of its broad application as solvent for Li–O₂ batteries to enable reversible oxygen reduction and evolution ($2\text{Li} + \text{O}_2 \rightleftharpoons \text{Li}_2\text{O}_2$), DMSO can be decomposed to dimethyl sulfone, making it not suitable for the development of long life cycle Li–O₂ batteries.⁵³

It should be noted that so far Li–O₂ batteries have been designed to enhance the delivered capacity and life cycle, leaving aside circular economy concepts such as resource and energy efficiency, material toxicity, process simplicity, or recyclability.⁵⁴ The multicomponent character and multistage fabrication of Li–O₂ batteries enhances its environmental impacts. Therefore, to obtain Li–O₂ batteries with reduced environmental impacts, sustainable one-pot processes which are energetically efficient and are atom-economic⁵⁵ or the use of earth-abundant materials with low toxicity is encouraged.

To shed further light on the distribution of environmental impacts within the analyzed batteries, the relative contribution of each battery component to the GWP is summarized in Figure 6. GWP is generally used as the most representative environmental impact, allowing cross-comparison with other energy storage systems.^{20,24,44,56} Indeed, CO₂ production is considered one of the “environmental key performance indicators”. GWP, measured in CO₂ equiv and defined as the

Table 2. Environmental Impacts for the Seven Studied Li–O₂ Batteries^a

Impact category	Porous carbon	MnO ₂ /graphene	Ru/graphene	3D Co/MOF	N-graphene	CoCO ₃ /Ag	Au/Ni/Ni	Li–O ₂ (avg)	Li–S (avg)	LIB Ecoinvent	NIB	units
fine particulate matter formation	58.34	55.15	92.23	42.57	83.78	300.17	238.10	124.34	311.68	349.70		× 10 ⁻³ kg PM 2.5 equiv
fossil resource scarcity	29.04	28.55	32.30	21.65	48.28	82.34	95.87	48.29	143.16	58.91	134.45	kWh equiv
freshwater ecotoxicity	6.90	6.71	4.87	4.64	8.91	17.45	80.90	18.63	6.82	106.77		kg 1,4-DCB
freshwater eutrophication	28.39	27.36	22.80	19.36	31.22	58.05	177.63	52.11	45.64	145.35	310.00	× 10 ⁻³ kg P equiv
global warming potential	30.61	29.32	40.24	23.67	41.98	121.91	102.57	55.76	130.57	58.43	146.35	kg CO ₂ equiv
human carcinogenic toxicity	3.25	3.29	2.36	76.69	3.45	4.56	10.57	14.88	8.65	10.28		kg 1,4-DCB
human noncarcinogenic toxicity	98.90	95.90	78.19	66.29	125.17	300.06	1153.04	273.93	96.32	1,092.23		kg 1,4-DCB
ionizing radiation	72.90	70.09	49.14	47.98	60.40	73.25	210.18	83.42	283.40	41.39		× 10 ⁻¹ kBq Co-60 equiv
land use	33.86	32.56	500.70	25.05	42.81	2427.65	244.39	472.43	26.62	61.67		× 10 ⁻² m ² a crop equiv
marine ecotoxicity	90.72	88.22	59.66	60.91	117.11	230.05	1061.77	244.06	87.69	1350.80		× 10 ⁻¹ kg 1,4-DCB
marine eutrophication	27.23	26.02	339.07	21.58	32.65	1,602.7	74.67	303.43	100.23	35.94	3100.00	× 10 ⁻⁴ kg N equiv
mineral resource scarcity	13.53	14.25	7.05	6.73	12.63	121.23	31.13	29.51	37.18	18.96		× 10 ⁻² kg Cu equiv
ozone formation, human health	7.42	6.99	12.01	5.70	9.57	43.12	33.46	16.90	41.62	24.08		× 10 ⁻² kg NO _x equiv
ozone formation, terrestrial ecosystems	7.53	7.09	12.44	5.79	9.85	44.65	33.90	17.32	42.78	24.76		× 10 ⁻² kg NO _x equiv
stratospheric ozone depletion	16.83	15.00	221.33	82.83	19.67	1020.3	557.67	276.24	194.90	45.20		× 10 ⁻⁶ kg CFC11 equiv
terrestrial acidification	13.09	12.26	27.44	9.68	19.74	99.07	55.71	33.85	78.03	91.05	131.38	× 10 ⁻² kg SO ₂ equiv
terrestrial ecotoxicity	67.37	62.40	102.32	53.46	146.43	334.17	260.33	146.64	309.36	5,313.06		kg 1,4-DCB
water consumption	34.37	33.59	23.36	22.58	35.31	35.29	101.31	40.83	114.32	50.35		× 10 m ³
global warming potential (GWP100a) CML-baseline	30.12	28.83	36.99	23.09	41.22	107.30	100.70	52.61	127.37	57.07	140.33	kg-CO ₂ equiv
terrestrial acidification potential (TAP) CML-baseline	15.90	14.90	28.76	11.63	22.72	97.68	67.67	37.04	95.73	109.32	151.00	× 10 ⁻² kg SO ₂ equiv
GWP DIF, CML/ReCiPe	1.65%	1.70%	8.79%	2.49%	1.86%	13.62%	1.86%	5.99%	2.51%	2.40%	4.29%	
acidification DIF, CML/ReCiPe	-17.70%	-17.74%	-4.61%	-16.73%	-13.10%	1.42%	-17.69%	-8.60%	-18.49%	-16.71%	-12.99%	

^aData corresponding to LIB is obtained from Ecoinvent 3.7 database. Li–S battery impacts are extracted from a recently published manuscript ref 24.

warming influence over a 100 year time horizon relative to that of carbon dioxide, is the major indicator for environmental arising from key anthropogenic greenhouse gases such as CO₂, CH₄, or N₂O. During Li–O₂ battery production, the major contributors to those gases are the burning of fossil fuels for electricity generation (CO₂), mining activities to extract raw materials (CH₄), and transport and combustion processes (N₂O).

On average, the cathode and the electricity required for material transformation and battery assembly are the most relevant drivers for the GWP, contributing by 44.5 and 17.0%, respectively, of the whole impact in this category (see the average of all seven batteries in the right part). Electricity consumption has been recently highlighted by Dai et al. as one of the barriers for the development of automotive batteries (for a study based on a lithium–nickel–manganese–cobalt oxide cathode LiNi_{1/3}Mn_{1/3}Co_{1/3}O₂, NMC111).⁴² Module packaging, composed by aluminum, steel, and copper wire, also has a relevant role as it contributes 15.6% of the whole GWP burdens because it requires significant amounts of aluminum and an anodizing process. The anode solely contributes to the 10.3% of the average GWP, while the electrolyte accounts for 3.1% of the GWP. Such marked contribution to the GWP of the electrolyte is in notable contrast with the recent results reported for Li–S batteries, where the electrolyte is the dominant contributor to the GWP with a relative weight of 35–47%.²⁴

The cathode solely contributes ~5% of the GWP of porous carbon battery, indicating a rather adequate design (environmentally speaking) of such battery as the active components do not substantially contribute to the GWP (anode contribution of ~18%). On the contrary, the relative contribution of the CoCO₃/Ag cathode to the GWP is of 83%, which should be reduced in the future taking into account the high emission rate of this type of battery (121.91 kg-CO₂equiv·kWh⁻¹). This research spots that in future developments, the cathode needs to be environmentally improved since it contributes to the 44.5% of the GWP in the studied Li–O₂ batteries, similar to the 39.1% greenhouse gas emissions reported for a NMC111 cathode.⁴² However, it should be taken into account that results are in lower than the 75% contribution to the climate impact of other Li–O₂ cathodes (cobalt oxide nanoparticles + cobalt(III) oxide + carbon nanotubes + PVDF binder).²² These differences clearly indicate the need of comprehensive studies covering diverse battery configurations to obtain representative average values.

Comparison with Lithium Ion and Lithium–Sulfur Batteries. A comparison between other battery technologies based on lithium provides the bigger picture on the possibilities of Li–O₂ batteries as sustainable energy storage systems. In this context, LCA results obtained in this research have been compared with the environmental impacts of a reference LIB from version 3.7 of the Ecoinvent database,¹⁰ a recently published study covering the cradle-to-gate LCA of lithium–sulfur batteries (the average of five Li–S batteries is shown),²⁴ a cradle-to-gate LCA of NIB, all of them having 1 kWh of storage capacity as a FU. As shown in Table 2, Li–O₂ batteries can potentially improve the environmental performance of existing battery technologies. The average environmental impacts of Li–O₂ batteries remain below the average impacts recently reported for Li–S batteries in terms of fine particulate matter formation, fossil resource scarcity, global warming, ionizing radiation, mineral resource scarcity, ozone

formation, terrestrial acidification, terrestrial ecotoxicity, and water consumption (9 impact categories out of 18). On the contrary, larger impacts regarding freshwater eutrophication, human toxicity (both carcinogenic and noncarcinogenic), marine ecotoxicity, marine eutrophication, and stratospheric ozone depletion are observed. Additionally, on average, Li–O₂ batteries require a much larger land use than do Li–S batteries (472.4 vs. 26.6). Greater environmental impacts have been reported in other 9 categories out of 18. These larger environmental impacts arise mainly from the use of scarce and highly toxic materials in the Li–O₂ cathodes, which are optimized to for high ORR and OER efficiencies, leaving behind circular economy principles.^{9,26} Furthermore, the average environmental impacts of Li–O₂ batteries are comparable to those corresponding to LIBs in terms of GWP, mineral resource scarcity and ozone formation, while Li–O₂ batteries beat the impacts of LIBs associated with fine particulate matter formation, freshwater ecotoxicity, human noncarcinogenic toxicity, terrestrial acidification, and terrestrial ecotoxicity. Those results are in line with the conclusions recently drawn by Want et al., who argued that a Li–O₂ battery aimed to sustain a middle-sized EV showed lower life cycle environmental burdens than a conventional LIB having a NMC cathode and a graphite anode.²³

It could be thus stated that, in general terms, Li–O₂ batteries present a slightly lower environmental impact than Li–S batteries, which shows a greater level of technological maturity (Li–S will reach commercialization soon, while Li–O₂ will not be commercially available before 2030). With an average value of 55.76 kg-CO₂ equiv, the relevant impact contributors such as GWP are 58 and 20% below the impact showed by LIBs and NIBs. More importantly, these impacts are expected to markedly decrease taking into account the early stage development of Li–O₂ batteries. The negligible amount of the so-called “dead weight” present in Li–O₂ batteries contributes to such low GWP values as it allows the development of notably lighter batteries for the same energy density (73–80% lighter batteries than Li–S or LIB). Interestingly, Li–O₂ batteries present considerably lower toxicological risks than do LIBs. For example, the average Li–O₂ human noncarcinogenic toxicity and marine ecotoxicity are 273.9 kg 1,4-DCB and 244.1 × 10⁻¹ kg 1,4-DCB, respectively, versus the 1092.2 kg 1,4-DCB and 1350.8 × 10⁻¹ kg 1,4-DCB for the reference LIB, respectively. Such lower toxicological risks may arise from the differences on the cathode and current collector materials. Indeed, LIB cathodes typically comprise active hosts for Li⁺ intercalation such as lithium cobalt oxide (LiCoO₂) or lithium manganese oxide (LiMn₂O₄),⁵⁷ together with a petroleum-derived polymeric binder and hard carbon derived from biomass carbonization. These cathodes are based on heavy and toxic metal oxides and require large amounts of water/energy/nitrogen (for carbonization) and nonrenewable materials,⁵⁸ markedly enhancing the environmental footprint of current LIBs. Additionally, LIBs use copper as a current collector, which markedly contributes to ecotoxicity indicators in comparison with the aluminum used by Li–O₂ technology.²⁰

We finally compared the environmental impacts obtained through the ReCiPe 2016 model with those arising from CML-baseline method. Due to the different impact categories in both methods, the comparison was only carried out for GWP and terrestrial acidification potential (see lower part in Table 2). This comparison has been also used to standardize the impact

values calculated for NIB batteries from CML to ReCiPe. We found that the ReCiPe 2016 model increases GWP by ~6% (average value for Li–O₂), while it provides lower impacts in the terrestrial acidification potential category (8.6%). This discrepancy also applies for reference LIB, NIB, and Li–S batteries and arises from differences in the sensitivity to each category.²⁷ However, we estimate that the results are reproducible as obtained differences remain below 10%. Both applied methods are updated versions of “ReCiPe 2016 v1.1” by RIVM, Radboud University, Norwegian University of Science and Technology, and PRé Consultants and “CML v4.7 August 2016” by Leiden University in The Netherlands.

Thanks to their lightweight character and high energy density, Li–O₂ batteries display decreased environmental impacts than other battery technologies per 1 kWh. However, the scarce/toxic materials within the Li–O₂ cathodes markedly contribute to enhance certain impact categories. As research on Li–O₂ batteries progresses and a higher level of maturity is reached, the use of critical raw materials should be replaced by abundant/nontoxic components, enhancing Li–O₂ battery sustainability.

Future Improvement Potential. To enable the practical implementation of Li–O₂ batteries and catalyze the transition to a cleaner energy based on circular economy principles, future efforts should concomitantly be aimed at enhancing the energy density and reducing the environmental impacts of Li–O₂ batteries. On the cathode side, fabricating oxygen cathodes which reduce the pore clogging effect during cycling should be a priority. If this could be done using simple synthetic approaches and renewable/abundant materials, then the obtained Li–O₂ batteries would present a reduced environmental impact. Cathodes composed by carbonaceous hierarchical hollow structures offer improved properties in comparison with nonhollow structures as they are able to buffer the effect volume variations upon charge/discharge.⁵⁹ Cathodes based on redox polymers (polymers containing groups that can be reversibly reduced or oxidized) represent a plausible solution to replace the inorganic and often scarce battery cathode as they can be obtained from low-cost abundant chemicals.⁶⁰

In contrast, the replacement of the lithium metal anode by lithiated carbon or aluminum⁶¹ would provide novel anode materials with improved stability in electrolytes while reducing the amount of required raw lithium, which contributes to environmental impacts mainly through waste generation, water contamination, and transport during mining (*ozone formation, fossil resource scarcity, mineral resource scarcity, land use, and ecotoxicity impacts*).⁶² Some lithium-containing salts (used as additives in electrolytes) such as LiTFSI show good chemical and thermal stability, so their use is recommended as they can be recovered and recycled. Similar to other battery chemistries such as zinc ion batteries,⁶³ the implementation of aqueous Li–O₂ batteries which do not require organic/toxic solvents such as TEGDME and would thus represent another worthy step toward sustainability. Finally, to ensure long-lasting batteries and therefore reduced environmental impacts, lithium dendrite formation should be prevented. In this field, gel-like polymeric electrolytes based on renewable resources which show a compromise between favorable Li⁺ transport characteristic of liquids and mechanical strength characteristic of solids⁶⁴ should be a priority.

Although Li–O₂ represents one of the most widely investigated battery chemistries within the metal–oxygen

category, recent advances in the field of energy storage suggest that sodium–oxygen batteries (Na–O₂), analogous to Li–O₂ in many aspects, can emerge in the future as an alternative with lower charge overpotential and reduced environmental footprint.⁶⁵ The substitution of the scarce lithium by the more abundant sodium (both as the anode and as salts in the electrolyte) would lead to cleaner and safer battery chemistries. Additionally, Na–O₂ batteries could use aluminum current collectors in comparison with the copper or nickel required for lithium-based batteries. The substitution of such materials contributes to develop lighter, cheaper, and recyclable batteries.⁶⁶

Battery recycling plays an important role in the global sustainability of batteries. Currently, material recovery is highly conditioned by existing technologies, the cost of recycling processes and the price of raw materials. The economic feasibility of current recycling technologies is controversial, and they are mainly focused on the recovery of valuable metals such as Co, Cu, Ni, Mn, Fe, or Al. Lithium is not considered in most of the processes, though its increasing scarcity is due to the expected larger demand. The same happens with the electrolyte composed of volatile organic elements, which due to its high toxicity should be a recycling priority, despite its relatively low importance in the overall weight of the battery.⁶⁷ After a mechanical pretreatment, two main technologies are applied: the pyrometallurgical process or combined pyrolysis at high temperature (metal reduction, ≈1500 °C) and medium temperature (gas incineration, ≈1000 °C) and the hydrometallurgical process, where the active cathodic powder is leached out to separate and purify the valuable metals. Each approach has advantages and disadvantages regarding material recovery and efficiency from the viewpoint of environmental impacts.²¹ A detailed information on these processes around the globe is found elsewhere.^{21,67}

As current recycling technologies for LIB electrodes are still in their early stages, in particular regarding the industrial exploitation of EV battery recycling (lack of maturity of technologies and nonexistence of a market for EV batteries in an end-of-life scenario), it is difficult to detail technologies related to the recycling of Li–O₂ battery components. However, we estimate that in the journey toward a circular economy, new recycling processes should increase the recovery rate of raw materials used in Li–O₂ batteries. Treatments with the highest recovery efficiency and the lowest environmental impact should be developed, allowing for raw material savings, economic gains, reduced energy consumption, waste minimization, and safe management of hazardous components.²¹ Further research work needs to be carried out in the near future to address the recovery and prospective reuse of catalytic particles within the opened Li–O₂ cathode, which are often based on highly toxic and scarce compounds. The use of separators based on polymers undergoing selective depolymerization back to their monomers would enable closed-loop life cycles toward a circular economy.^{68,69}

Other progress in the field may arise from the development of transient Li–O₂ batteries, which can be disintegrated into harmless byproducts, avoiding environmental impacts arising from the accumulation of current electronic/battery in the environment.⁷⁰ Thereby, human and environmental exposures to the hazardous pollutants present in conventional Li–O₂ batteries will be avoided, lowering impacts in the categories of freshwater ecotoxicity, human toxicity, marine ecotoxicity, and terrestrial ecotoxicity. Overall, some promising routes toward

the reduction of the environmental impact of Li–O₂ batteries comprise the replacement of inorganic and scarce materials used in cathodes by renewable/abundant compounds, the use of lithium alloy anodes, the pursuit of alternative metal–oxygen chemistries, the development of degradable/transient batteries, and the design for recycling.

CONCLUSIONS

Here we tap into the possibilities of Li–O₂ batteries as an emerging energy storage technology with potentially reduced environmental impact. A comparative cradle-to-gate life cycle assessment analysis per 1 kWh of storage capacity on seven Li–O₂ batteries presenting cathodes of different characteristics is performed. To enable future cross-comparison, the life cycle inventories of batteries showing energy and material inputs are disclosed. Environmental impacts are assembled into 18 standardized ReCiPe 2016 Midpoint (Hierarchical) impact categories. Batteries comprising a cathode based on 3D Co/MOF, porous carbon, and MnO₂/graphene show reduced environmental impacts in comparison with cathodes based on CoCO₃/Ag and AuNi/Ni. With contributions of 44.5 and 17.0% on average, the cathode and the electricity required for material manufacturing and battery assembly are the most relevant drives for global warming potential, respectively.

Li–O₂ batteries present an average value of 55.76 kg-CO₂ equiv in GWP, 472.43×10^{-2} m² a crop equiv in land use, and 18.63 kg 1,4-DCB in freshwater ecotoxicity. A comparison with lithium ion batteries, Li–S batteries, and sodium ion batteries reveals that Li–O₂ batteries offer an enormous potential to reduce the environmental impacts of existing battery technologies, especially those related to toxicological risks. Such reduced impacts are mainly related to the simplicity/efficiency of the cathode fabrication, the use of abundant and safe materials, and limited amounts of electrolyte. Atom-economic and energy-saving fabrication technologies such as one-pot processes may also provide reduced environmental impacts. In the near future, the progression toward a circular economy will be a social and environmental necessity, while further follow-on works in the field would allow a cradle-to-cradle LCA analysis, providing a better insight on the environmental impacts. In the meantime, the results here reported provide a promising trail head to develop novel sustainable energy storage systems, in particular given the low technical maturity of Li–O₂ technology.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acssuschemeng.1c01554>.

Tables summarizing the inventory modeling of battery materials production (PDF)

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Author Contributions

The manuscript was written through contributions of all authors. All authors have given approval to the final version of the manuscript.

Notes

The authors declare no competing financial interest. All the data used to support the findings of this study are included within the article.

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