The new-type batteries with ultimate energy density

Ming He1,a,†, Maoxun Wang2,b,*,† and Zerui Wang3,c,†

1Research Institutes of Leather and Footwear Industry of Wenzhou, Wenzhou 325005, China
2Department of B-tech Automotive, McMaster University, 1280 Main St W, Hamilton, Canada
3Institute of New Energy and Low-Carbon Technology, Sichuan University, Chengdu 610207, China
aguanghua.ren@gecademy.cn; b*wangm121@mcmaster.ca; c1154213579@qq.com
†These authors contributed equally.

Abstract. In recent years, many countries have made plans for the development of electric vehicles. In 2021, the EU announced a plan to completely stop the sale of fuel cars by 2035 and replace all fuel cars with pure electric vehicles, reducing the carbon emissions to 100%. This is the most radical emissions reduction plan in history, and it means that the era of pure electric cars has officially arrived. The limited energy density of lithium-ion batteries currently used in cars has hampered the development of electric vehicle mileage. To meet the demand for electric vehicles, the development and research of high energy density batteries are urgent. Based on a review of the current literature, this paper summarizes the development history, working principles, current challenges and solutions of the solid-state battery, lithium-air batteries and nuclear batteries. The current dilemma for solid-state batteries is the lack of a suitable solid electrolyte, which is needed to possess high ionic conductivity of above 10 (mS/cm) at room temperature and negligible electronic conductivity with a high ionic transference number wide electrochemical stability windows. Lithium-air batteries have low power density, battery energy attenuation, and high safety performance. The research and application of nuclear batteries are more difficult, including low energy conversion rate and health problem. The result provides some guidance to researchers initially involved in the high energy density battery industry.

1. Introduction
Nowadays, fossil energy in the world is increasingly exhausted, which leads to the increasing demand for sustainable energy with zero-carbon emissions. Many countries have specified strict measures to promote the development of a low-carbon economy. The Chinese government has issued a development plan for the electric vehicle industry. In the next 15 years, the electric vehicle industry of China will adhere to the development direction of electric, connection, and intelligence. By 2025, the average power consumption of pure-electric vehicles will be reduced to 12 kWh per 100 km, and the sales volume of electric vehicles will reach about 20% of the total sales volume of new vehicles. Besides, the UK government's "Road to Zero" strategy calls for at least 50 percent of cars and 40 percent of vans in the UK to have ultra-low emissions (CO2 below 50 g per km) by 2030. The government plans to ban the sale of new cars powered by traditional internal combustion engines by 2040. However, current lithium-ion batteries cannot fully meet the needs of electric vehicles, especially in the driving range. The energy
density of conventional lithium batteries is difficult to be improved due to the limitation of the electrode and electrolyte materials. This has created an urgent need for cheap, environmentally friendly and efficient energy storage equipment, especially in the context of the rapid development of electric vehicles in recent years. At present, three ideal types of batteries can provide high energy density in theoretical, solid-state batteries, lithium-air batteries, and a special kind of battery, nuclear batteries. This paper was divided into three sections covering three types of high energy density batteries, including solid-state batteries, lithium-air batteries and nuclear batteries. Then this paper describes the working principle, research status, advantages, challenges and solutions of three types of batteries. It has guided the researcher who is interested in high energy density batteries.

2. Solid-state battery

2.1. Background
Solid-state batteries (SSE) have been around for a long time, dating back to Michael Faraday’s study on the fast solid-state ion PbF$_2$ and Ag$_2$S in 1838. Solid-state batteries were introduced as commercial batteries in 2010, but the concept of SSE dates back 200 years to Michael Faraday's work on fast solid-state ion transport in PbF$_2$ and Ag$_2$S[1]. Solid-state battery materials with practical value have emerged since 1960, mainly divided into the inorganic solid-state battery (SIEs) and polymer solid-state batteries (SPEs).

At present, the inorganic solid electrolytes are perovskite, NASICON, garnet and sulfide materials. The most mentioned perovskite electrolyte is Li$_{1-x}$La$_{2/3-x}$TiO$_3$, which exhibits lithium-ion conductivity excess $10^{-3}$ S cm$^{-1}$ at room temperature [2]. NASICON generally refers to Na$_{1+x}$Zr$_2$Si$_x$P$_{3-x}$O$_{12}$ developed in 1976[3]. The general expression of garnet-type material is A$_3$B$_2$Si$_3$O$_{12}$. Li$_{6.5}$La$_3$Zr$_{1.75}$Te$_{0.25}$O$_{12}$ achieves high ionic conductivity of $1.02 \times 10^{-3}$ S cm$^{-1}$ at room temperature, considered solid electrolyte material with great application potential[4]. Sulfide solid electrolytes, generally referred to as Li$_2$S-SIS-type electrolytes, were first studied in 1986[5].

There are three types of polymer electrolytes for lithium batteries: dry solid polymer electrolytes, gel polymer electrolytes, and composite polymer electrolytes. Polymer batteries are usually ceramic materials filled with a polymer body to form a new composite polymer electrolyte.

2.2. Working principle
In essence, the working principle of solid-state batteries is similar to conventional lithium-ion batteries, which rely on lithium ions moving back and forth between positive and negative electrodes to charge and discharge. The difference is that solid-state batteries are consist of solid electrolytes, whereas conventional lithium-ion batteries consist of liquid electrolytes.

Schottk y and Frenkel's defects promote ion transport in SIE, and the rate of ion transport depends on the concentration and distribution of the defects. As shown in Fig.1, an interfacial layer is formed when two materials with different chemical properties come into contact. This interfacial layer also affects the rate of ion transport[6].

![Fig.1 Ion transport in SPEs and SIEs (a) Schematic diagram of conduction principle of SIE ion (b) Schematic diagram of conduction principle of SPE ion][7]
The ion transport rate in polymer electrolytes is related to the glass transition temperature and the piecewise motion of the polymer chain[7]. In response to an electric field, lithium ions hop from site to site in a polymer chain, transferring ions by successive jumps. This ion transport capacity depends on the number of free lithium ions in the polymer.

2.3 Challenges
The advantages of SIE are generally high ionic conductivity ( >0.1 mS cm\(^{-1}\) at room temperature), high modulus (> 1 GPa), wide electrochemical stability window ( >4.0 V) and excellent thermal stability (stable at 100 °C)[8]. However, SIE is still difficult to get practical application because SIE has the following major challenges. It is necessary to solve the problem of material brittleness in a large area when SIE is faced with large-scale commercial manufacturing. In addition, SIE is prone to poor contact between solid and solid surfaces compared with liquid electrolyte batteries, which leads to poor interface charge transfer. Metal dendrites may also grow along grain boundaries during the charging and discharging of batteries. Finally, SIE materials also face the problem of high cost in the process of commercialization.

SPEs have some advantages: simple synthesis, low density, stable chemical properties, low cost, and easy large-scale manufacturing. However, many polymer materials (polyethylene and polypropylene) have low dielectric constants, which causes ion pairs in the electrolyte to be difficult to dissociate. In this case, cations can be difficult to transport.

2.4 Solutions
Solid electrolyte materials need to meet high conductivity, good electrochemical stability and acceptable mechanical properties. A relatively reliable approach is to study various complex problems of ion transport mechanisms through the use of computer modeling, the use of integrated methods through experiments, and the use of advanced characterization techniques. To achieve rapid ion conduction, the structure must meet three minimum criteria. The number of equivalent (or nearly equivalent) sites available for mobile ions should be much greater than the number of mobile species. The migration barrier energy between adjacent available sites should be low enough so that ions can easily jump from one site to another. And these available sites must be connected to form a continuous diffusion path[9-10].

SPEs further study the chain's segmental movement, which can produce a free volume for the hopping of lithium ions in coordination with polar groups. With the segmental movement of the polymer chain, lithium ions jump from one coordination site to another[11-13].

3. Lithium-air battery

3.1 Background
The lithium-air battery is a kind of battery that uses lithium metal or materials containing lithium as the negative electrode, oxygen as the positive electrode. It uses non-aqueous organic electrolyte or asymmetric electrolyte coexistence of water and non-water system or water-electrolyte or solid electrolyte as the working electrolyte.

Lockheed first proposed Lithium-air batteries in 1976. A metal lithium-air battery consists of of an alkaline aqueous solution as an electrolyte. The lithium-air battery has attracted widespread attention since it was proposed because of its high energy density. In 1996, Abraham and Jiang proposed a lithium-air battery based on a polymer electrolyte/organic electrolytic liquid system, solved the problem of lithium negative electrode reacting with water, and opened a new stage in the lithium-air battery research [14]. The Department of Energy's Vehicle Technologies Office (VTO) launched the Innovation Centre for Battery500 Consortium in 2016. By implementing the innovations developed under the Battery500 Consortium, a record of 600 stable cycles has been demonstrated in a prototype 350 Wh kg\(^{-1}\) lithium-metal pouch cell. The consortium has also developed a 400 Wh/kg lithium-metal pouch cell and now is pushing toward 500 Wh kg\(^{-1}\)[15]
3.2 Working principle
As shown in Fig.2, a lithium-air battery is a chemical battery with metal lithium as the anode, oxygen adsorbed on porous carbon as the cathode and an organic electrolyte system. In the discharging, lithium metal is oxidized to lithium ions into the organic electrolyte. The oxygen dissolved in the cathode into the electrolyte and was reduced to O$_{2}^{2-}$ under the action of catalyst, and when combined with the lithium-ion in the electrolyte to deposit Li$_2$O$_2$ on the cathode surface. The charging process is the reverse of this process. Note that some authors have reported that discharge down to Li$_2$O is possible, which would increase the energy stored but may be difficult to reverse on a charging[16-18].

![Fig.2 Schematic representations of Li-air battery (non-water)[19]](image)

Lithium-air batteries have high energy density and specific energy due to their special structure and electrode materials derived from the air. As shown in Table 1, compared with current lithium-ion batteries, lithium-air batteries have obvious advantages in energy density. At the same time, the battery has a wide range of material sources and little environmental pollution.

| Theoretical specific energy (Wh kg$^{-1}$) | Theoretical energy density (Wh l$^{-1}$) |
|------------------------------------------|----------------------------------------|
| Today's Li-ion ($\frac{1}{2}$C$_6$Li + Li$_0.5$CoO$_2$) | 387 | 1,015 |
| Li–O$_2$ (non-aqueous) | 3,505 | 3,436* |

3.3 Challenges
The main challenges of lithium-air batteries today are poor cycling and poor charge-discharge performances. The root cause of these challenges is the cathode and anode material of the battery. Fig.3 is a schematic of the cathode. An oxygen permeable membrane (OPM) layer filters carbon dioxide and water from the air and allows oxygen to pass through. When the oxygen passes through the OPM layer to the gas diffusion layer (GDL) and then through the gas diffusion to the catalyst layer (CL), it is catalyzed and finally reduced in the electrolyte.
3.3.1 CO2 and H2O from the air. The presence of carbon dioxide will incur neutralization of the alkaline electrolyte and precipitation of carbonate that blocks the transport channels. The inclusion of water will cause Li anode corrosion and electrolyte degradation. To address these problems, it is very helpful to use a membrane that is O2 permeable but can block the incorporation of CO2 or H2O from the air[20].

3.3.2 Sluggish kinetics. Since the active material of the cathode in the lithium-air battery is gaseous oxygen, the electrochemical reaction needs to be dissolved in the electrolyte solution first and then get electrons from the phase interface to be reduced. The reduction process also needs the help of a catalyst, so from the perspective of chemical kinetics, their reaction is very slow. The electrochemical kinetics can be improved effectively by developing the catalyst with higher performance and adjusting the electrode structure and electrolyte parameters (concentration, conductivity, composition).

The catalyst materials can be divided into four types: Noble metals and alloys, Carbonaceous materials, transition-metal oxides and Inorganic-organic composites. At present, Noble metals are the catalyst with the best performance and the highest cost, while transition-metal oxides are slightly cheaper materials with the most potential to replace conventional catalysts. In recent years, the lithium metal anode has been widely concerned by researchers because of its high theoretical capacity, low potential, and good electrical conductivity. It is also one of the most promising anode materials. However, lithium anodes also have some disadvantages that are difficult to be overcome.

3.3.3 Dendrite growth. The dendritic deposition is a common phenomenon in the high-current electroplating process of metals such as Cu, Ni, and Zn, but this phenomenon has been fully researched and has no effect on industrial applications[21].

![Fig.3 proposed electrode structures][20]

Fig.3 proposed electrode structures[20]

Fig.4 shows the Li stripping/plating process, which consists of four steps. Step 1. Lithium plating causes volume expansion, causing cracks in the solid electrolyte interphase (SEI) film. Step 2: Further plating causes lithium dendrites to be ejected from the cracks. Step 3: Li stripping produces isolated Li, which becomes part of the "dead" Li, and the volume shrinkage causes the SEI to break further. Step 4: Continuous cycling causes steps 1-3 to happen repeatedly, which ultimately leads to accumulated dead lithium, thick SEI, and porous lithium electrodes[22].

As a result, this will lead to the capacity fading. At the same time, the growth of lithium dendrite can penetrate the membrane and cause a short circuit inside the battery, which leads to serious safety problems. At present, many researchers have solved the problem of dendrite from electrolyte and interface.
3.4 Solutions
It is found that the use of different electrolyte additives has a huge impact on the formation of SEI. For example, a study of HF shows that a small amount of HF and water in the carbonate electrolyte can generate a uniform and dense LiF/Li₂O layer on the Li surface[23]. This dense passivation layer can play an effective protective role, resulting in relatively smooth hemisphere deposition of Li. But this does little to help the Coulomb efficiency and will disappear after a few more cycles. At the same time, many researchers will do some artificial SEI to limit the dendritic problem. For example, on a clean Li surface, the reaction between the substituted silane and the natural OH terminal layer results in a stable, protective layer under static conditions, with low initial impedance and slow impedance growth when exposed to organic electrolytes[24].

4. Nuclear battery

4.1 Background
The study for the nuclear battery was started in 1913 when Henry Moseley published the technology or generated electrical power from the isotope. Scientists have put more attention on long-life battery technology to fill the demand. RCA worked on a tiny atomic battery for small radio receivers and hearing aids in 1954. And nuclear battery got rapid development science then[25].

4.2 Working principle
The nuclear battery is a device that generates electricity by using energy from the decay of a radioactive isotope. They create electricity from nuclear energy like nuclear reactors do, but they do not require a chain reaction. Although they are usually referred to as batteries, they are not electrochemical in principle and cannot be charged or recharged[26]. They are extremely expensive in comparison but have a very long life and high energy density, seen in Fig 5, so they are primarily used as energy sources for equipment that must operate unattended for extended periods.

![Fig.5 Energy Density of radioisotope battery producing electrical current][26]

Nuclear batteries are also called radioisotope batteries. Isotopic cells come in two types, one with a thermal converter and the other with a non-thermal converter[26]. Radioisotope batteries are made from long-lived isotopes of radioactive elements such as strontium-90 (Sr-90), with a half-life of 28 years, plutonium-238 (Pu-238), with a half-life of 89.6 years and polonium-210 (Po-210), with a half-life of 138.4 days. The principle of a heat converter isotope cell is to make it into a cylindrical cell. The fuel is...
placed in the center of the cell, and the radioisotope emits high-energy alpha rays. The heat generated by the radioactive elements is transmitted through a thermonuclear wire made of different metals, forming a voltage between the wires, seen in Fig 6[27]. The radioactive isotope battery used in the US lunar rover is plutonium-238, which has a rated power of 63.5 watts and a life span of one year. Radioisotope batteries are used to power the lunar rover because they operate in an environment unaffected by external factors[28].

Non-thermal conversion is one of the energy conversion methods of nuclear batteries. A generator absorbs radiation directly from the isotope before the particle is degraded into thermal energy. Due to the diversity of isotopes, a different method was demanded the conversion. There are 'Electrostatic conversion', 'Direct-charging generator', 'Electromechanical conversion', 'Radiovoltaic conversion', 'Alphavoltaic conversion', 'Betavoltaic conversion', 'Gammavoltaic conversion', and 'Radiophotovoltaic (optoelectric) conversion' [29]. Moreover, 'Betavoltaic' is the most commonly used technology due to the longer life span and lower radiation shielding, despite 'Alphavoltaic conversion' and 'Gammavoltaic conversion' creating more potential energy.

Betavoltaics are electric current generators that use energy from a radioactive source that releases beta particles. Tritium is a frequent radioactive source for Non-thermal conversion. To create electricity, they use a semiconductor p-n junction. When Beta emission is applied to an electrode, it produces electron-hole pairs, raising the potential between the electrode and the current flow[29].

4.3 Challenges
There are also many drawbacks of nuclear batteries, resulting in the situation that electric vehicles running today do not use nuclear power. Since the isotope has been produced, it has been decaying, and its electrical properties have decreased. First of all, the isotope plutonium-238 is extremely difficult to manufacture. As a nuclear power, the United States now produces only 1.5 kilograms of plutonium-238 per year. Compared with the several kilograms of plutonium-238 needed by a rover, the raw material is extremely rare.

On top of that, the radioisotope batteries on the rover cost 70 million dollars. It is impossible to use it on a car with that price. Also, the energy conversion rate is not high[25, 29]. The maximum ratio of the energy generated by the radioisotope is 6-8 %. The energy conversion rate of the nuclear battery made in China is only 0.1 %[28]. The most worrying point is that radioactive isotope batteries are radioactive. Using nuclear power in civilian cars will cause some serious safety problems, especially when an accident happens.

![Fig.6 Principle of radioisotope battery producing electrical current[30]](image-url)
4.4 Applications
The property of the raw material in nuclear batteries satisfied the requirement of deep space exploration. Applying the radioisotope battery refrains from the large expenditure of taking the primordial power source into space. The nuclear battery also consists of of some advantages, such as no need for recharging the batteries. Nuclear batteries power pacemakers. Pacemakers are medical devices that use electrodes to transmit electrical impulses to contract the heart muscles and control the heart's electrical conduction system [29].

5. Conclusion
Since the successful commercialization of lithium-ion batteries in 1991, they have been widely used in electronic equipment, electric vehicles, energy storage and other fields. However, the existing commercial lithium-ion battery system is difficult to continue to improve the energy density and inherent safety problems, causing the development of lithium-ion batteries has encounter a bottleneck. Using other types of batteries to achieve higher energy density is an effective way to solve this problem. In recent years, solid-state batteries, lithium-air batteries and nuclear batteries have attracted increasing attention as potential high energy density batteries.

Solid-state batteries using solid electrolytes can fundamentally solve the problem of lithium-ion battery safety and further improve the battery's energy density. However, the current Solid-state battery electrolyte also has some difficult problems, such as interfacial impedance and mechanical properties. Advanced theoretical calculations and computer-aided screening can greatly accelerate the discovery of electrolytes, especially for inorganic electrolytes with periodic structures. Although there are still errors in the structure of the calculation and the experiment, this calculation can quickly exclude many compounds with negative properties to reduce the size of the experiment.

The energy density of lithium-air batteries is higher than that of solid-state batteries, but the charge-discharge cycle efficiency is lower. Besides, dendrite growth is a difficult problem to solve. Researchers are currently trying to solve this problem by using electrolyte additives and making artificial SEI membranes.

As one of the ideal high energy density batteries, nuclear batteries are the most difficult in technology. Nuclear batteries have the advantage of unparalleled energy density. However, the intractable radioactive residues and serious safety issues cause nuclear batteries difficult to commercialize. At present, nuclear batteries are mainly used in space and some military fields, but nuclear batteries can be expected to find wider applications in the future.

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