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Review

Aluminum-Based Fuels as Energy Carriers for Controllable Power and Hydrogen Generation—A Review

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Abstract: Metallic aluminum is widely used in propellants, energy-containing materials, and batteries due to its high energy density. In addition to burning in the air, aluminum can react with water to generate hydrogen. Aluminum is carbon-free and the solid-phase products can be recycled easily after the reaction. Micron aluminum powder is stable in the air and enables global trade. Aluminum metal is considered to be a viable recyclable carrier for clean energy. Based on the reaction characteristics of aluminum fuel in air and water, this work summarizes the energy conversion system of aluminum fuel, the combustion characteristics of aluminum, and the recycling of aluminum. The conversion path and application direction of electric energy and chemistry in the aluminum energy conversion system are described. The reaction properties of aluminum in the air are described, as well as the mode of activation and the effects of the aluminum-water reaction. In situ hydrogen production is achievable through the aluminum-water reaction. The development of low-carbon and energy-saving electrolytic aluminum technology is introduced. The work also analyzes the current difficulties and development directions for the large-scale application of aluminum fuel energy storage technology. The development of energy storage technology based on aluminum is conducive to transforming the energy structure.

Keywords: recyclable energy carrier; aluminum fuel; aluminum combustion; aluminum-water reactions; energy storage; energy conversion system

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1. Introduction

Fossil energy will still be the main form of energy in the world for a long time in the future [1–6]. The long-term direct discharge of fossil fuels has caused a series of environmental problems. Meanwhile, the limited availability of fossil fuels forces countries to carry out the transformation of the energy structure [7–9]. There is an urgent need for a low-carbon fuel with high energy density that can work alongside fossil fuels to generate electricity and heat, power vehicles, and trade globally [1,10]. Among them, biomass, solar, wind, water, nuclear, and hydrogen are new and clean energy sources that have long received widespread attention and development [11–16]. The majority of the new energy sources are characterized by intermittency, instability, and uncertainty, which have severely restricted their large-scale development. Hydrogen with its heat of combustion value of 142,351 $kJ \cdot kg^{-1}$ is very high among the commonly used fuels [17]. Hydrogen combustion is very clean and the only product is water [15–17]. Hydrogen energy can be used in a broad range of industries, but the storage and transport of hydrogen have long been constraints to its further development [18,19]. Therefore, the key challenges of the current transformation of the energy structure are not only the development of clean energy, but also the development of large-scale, long-cycle storage technologies, as well as clean energy for global transportation and trade [20].

Energy storage mainly refers to the storage of electricity, which can be stored by converting it into chemical, potential, kinetic, and electromagnetic energy forms [20,21]. Energy storage technology has come a long way in recent decades [22]. The classification and application summary of common energy storage technologies are presented in Figure 1 [20–27]. As demonstrated in Figure 1, the various energy storage technologies have obvious differences in energy and power density. There is no one-size-fits-all technology that can meet all energy storage needs [25]. The significant progress in batteries has provided new ideas for the development of energy storage technology [28–30]. Batteries are hardly available for large devices and long distances, and their service life is controversial [1]. Chemical energy is the most reliable source of energy for society by far [27,31].

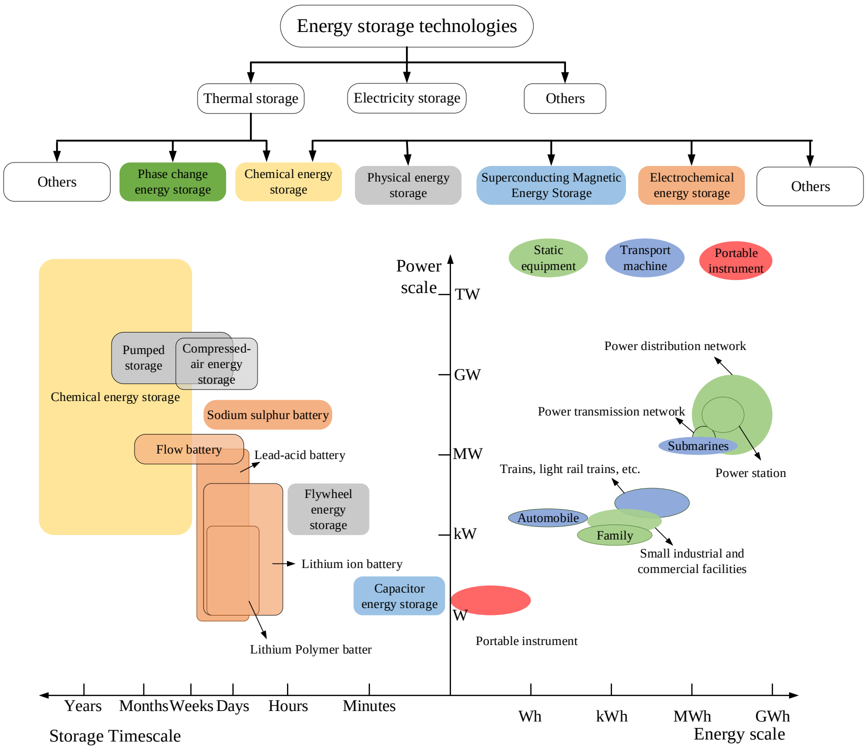


Figure 1. Common energy storage technologies and applications [20–27].

At present, there is no reliable substitute for energy trade, and it is difficult to replace fuel with energy storage technology in the market trade. Innovative technologies need to be developed to achieve multi-disciplinary synergies. In addition to the progress of energy storage technology, it is also urgent to find energy carriers to promote the comprehensive transformation of the energy structure [11,12,31]. Sound energy carriers need to have essentially the following characteristics in order to meet the energy needs of social development [27,31–33]:

1. Carbon-free or low-carbon throughout the life cycle;
2. High energy density;
3. Abundant, inexpensive, or recyclable reserves;
4. It allows for global trade and can be stored safely with little loss;
5. It can meet various power levels of transportation and fixed generation.

Figure 2 presents the properties of metallic fuels compared to common solid and liquid fuels [10,18,34–39]. According to Figure 2, hydrogen has a very high specific energy. However, even the density of liquid hydrogen is too low and hydrogen is extremely difficult to store and transport safely [18]. It is also clear from Figure 2 that metal fuels have the ability to be promising energy carriers.

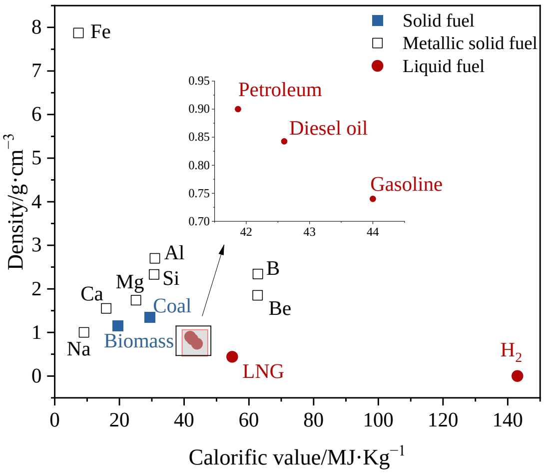


Figure 2. Comparison of the properties of common solid and liquid fuels [10,18,34–39] (LNG is Liquefied Natural Gas; compressed natural gas has a calorific value of 48 but its density is extremely high and is, therefore, not listed; there are many types of fuels such as coal and biomass, and their calorific values and densities are taken as average values).

In contrast to conventional fossil fuels, despite combustion reactions with air, some reactive metals are oxidized by water to generate hydrogen [8,40]. Figure 3 illustrates a schematic diagram of the metal fuel storage process [27]. Unlike conventional energy storage technologies, metal fuels and metal oxides achieve energy storage in the conversion process [27]. Metals have a long storage cycle and a small loss, enabling global trade. Figure 4 shows a summary of the properties of common reactive metals [11,40–44]. Figure 4 compares the heat of different metal–water systems, the volume of hydrogen produced by the complete metal–water reaction, and the amount of metal in the geological crust. Considering the energy density, the reaction with water to produce hydrogen and exothermic heat, storage conditions, and safety, it is concluded that aluminum has the potential to be a viable carrier for hydrogen storage and energy storage [32,44,45]. The aluminum metal has a high energy density, which is environmentally friendly, and its post-combustion products are recyclable [1,10,23,46]. Aluminum not only burns in air (oxygen) and carbon dioxide, but also reacts with water to produce hydrogen, while giving off a lot of heat. Most work on the aluminum-water reaction has focused on the hydrogen produced at low temperatures as an in situ hydrogen source [45]. The exothermic heat of the aluminum-water reaction at low temperatures is frequently wasted [1,10,27]. Although there are still some challenges in the large-scale application of aluminum fuel, the clean and efficient application of aluminum fuel has a broad prospect.

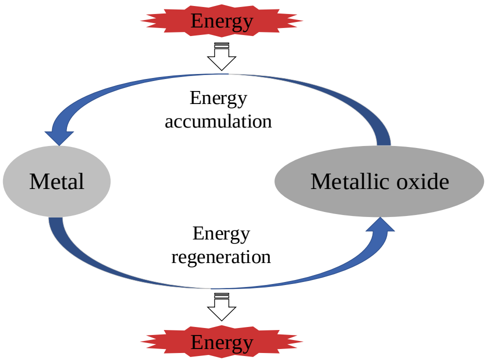


Figure 3. Schematic diagram of the metal fuel energy storage process [27].

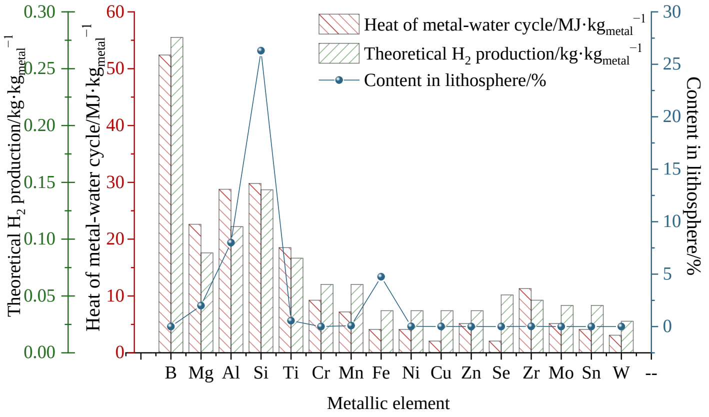


Figure 4. Comparison of the properties of common metal fuels [11,40–44] (hydrogen production: theoretical hydrogen production from the metal–water reaction; heat of reaction: the heat of circulation in the metal–water circulation system).

2. Aluminum-Fuel-Based Energy Conversion Systems

Aluminum is oxidized in air (oxygen), water, and carbon dioxide. The studies on Al-CO_2 are mainly conducted in deep space [47,48]. Therefore, this paper focuses on the mechanism and application of aluminum-air (oxygen) reaction and aluminum-water reaction. Aluminum fuel is applied in a wide variety of situations, such as underwater propellants, rocket propellants, explosives, fireworks, and batteries [37,44]. Hydrogen and heat are generated during the reaction of aluminum with water [1,10]. Hydrogen is frequently used as fuel for engines and fuel cells, and propellants for rockets and underwater vehicles commonly use the heat generated by the reaction [37,44]. Normally, propellants need to react at high temperatures ($T > 3000$ K) [37]. Currently, aluminum-water reactions for the preparation of hydrogen are generally performed at low temperatures (below the boiling point of water) [44]. The low-temperature reaction only utilizes the chemical energy in the hydrogen and the heat is seriously wasted. Propellants typically just utilize the exothermic heat of the reaction and the hydrogen produced is vented outward to generate power. Chemical energy is wasted in aluminum. Bai et al. [49] proposed an energy storage system based on aluminum-air reaction. The system has been thermodynamically evaluated by Aspen Plus and compared with other systems. The results show that aluminum-fueled energy storage systems have a higher roundtrip efficiency and that the cost of electricity from aluminum-fueled energy storage is comparable to that of coal-fired power plants.

This work focuses on energy conversion systems that allow the full exploitation of the chemical energy in aluminum-based fuel. Zero-carbon energy conversion systems at scale require high energy and power densities to enable the supply of power to households, industry and equipment, remote power sites, grids, etc. [1]. Similar to conventional hydrocarbon fuels, the combustion of aluminum powder at high temperature and pressure is required for the system to obtain high energy and power density. Depending on the oxidizer, aluminum-fuel-based energy conversion systems are classified into the following two pathways, as shown in Figure 5 [1,49].

(1) Aluminum-air reaction: Similar to fossil fuels, aluminum powder is combusted directly in air, releasing heat to fuel heat engines or for use in fuel cells.

(2) Aluminum-water reaction: Hydrogen and heat are obtained during the aluminum-water reaction, which allows its application in engines, heat engines, and fuel cells [45].

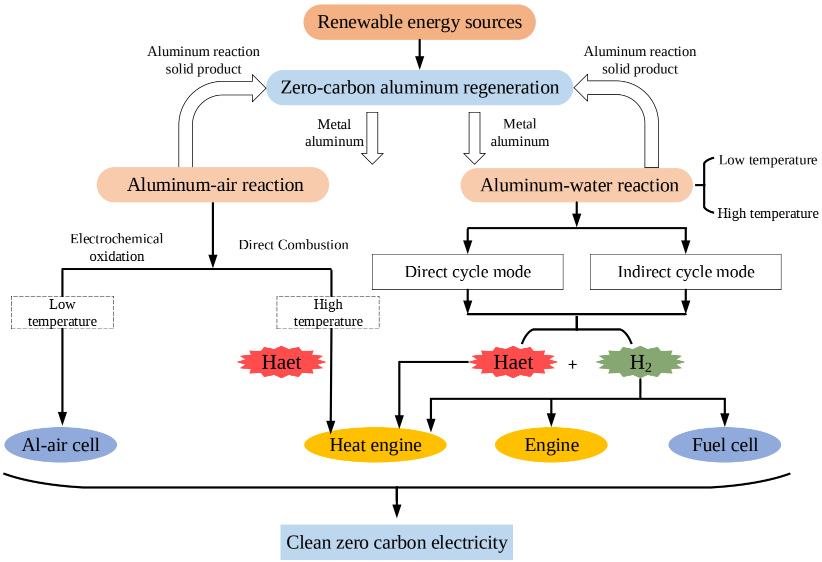


Figure 5. Schematic diagram of the zero-carbon-cycle power generation system based on aluminum fuel.

The two routes are disclosed in Figure 5. Each of these loops shows two paths. The aluminum-air reaction has both high- and low-temperature pathways. One route is the aluminum-air electrochemical reaction inside the cell at low temperatures. The other route is the combustion of aluminum powder in air at high temperatures, which has an energy density higher than that of the low-temperature route. Aluminum has a high chemical equivalent value, 2.98 $\mathrm{Ah \cdot g^{-1}}$, second only to lithium [50,51]. Aluminum-air batteries convert the chemical energy of aluminum directly into electrical energy [52]. Aluminum-air batteries mainly consist of an aluminum anode, electrolyte, and air electrode [51,53]. Aluminum-air batteries are environment-friendly in operation and also have the advantages of high specific energy, low mass, and low noise [52,54]. Aluminum-air batteries are highly adaptable and stable, but they have relatively low power, low electrochemical efficiency, and high self-discharge rates [55]. In addition to technical challenges, the high cost of electricity for aluminum-air batteries also restricts its large-scale application [56]. Numerous studies have concluded that aluminum-air batteries have great potential and advantages for applications such as underwater power supplies, electric vehicles, and power supply stations [55,57,58]. Electrochemical oxidation is also one of the pathways for the realization of aluminum as the energy carrier.

The aluminum-water reaction also occurs in both high- and low-temperature modes. The hydrogen and heat from the reaction are commonly used in various turbines and fuel cells [45]. Whatever the route in an aluminum-fuel-based energy conversion system, clean renewable primary energy is converted into the chemical energy in aluminum. Aluminum is easy to store and transport, with the subsequent application of aluminum fuel as required [37]. The solid products of the reaction are collected and can be regenerated into aluminum, which continues to participate in the system cycle. Regarding aluminum powder particle size, there are problems of incomplete combustion and loss of combustion efficiency with micron particles [1]. Nanometer aluminum powder avoids such problems [59]. However, nanoparticles are expensive, their energy recycling is low, and they have safety issues. Nanoparticles and its oxides may cause wear problems, and the separation as well as collection of both is difficult. In contrast, micron-sized aluminum powders offer higher energy density, cycle efficiency, and safety, and are less expensive.

2.1. Aluminum-Air Reaction Power Generation

Aluminum combustion in air directly provides the system with high power generation efficiency and energy density [37]. The combustion reaction equation is indicated in Equation (1).

$$4\text{Al} + 3\text{O}_2 = 2\text{Al}_2\text{O}_3 \tag{1}$$

The metal–air reaction is expected to apply in internal combustion engines to attain high utilization efficiency [1]. The same is true for aluminum powder. The development of specialized metal combustors for burning metals increases the viability and reliability of the system and is one of the pivotal points of current research. Bergthorson [10] proposed a principle for the utilization of systems based on the metal–air reaction, including a cyclone combustor that is usable for metals. This metal cyclone burner mainly consists of a metal fuel storage tank, a metal fuel combustion chamber, a combustion product cyclone separator, and a combustion product storage tank. The metal fuels are violently oxidized by the air in the combustion chamber. The solid products created by the reaction are separated and recovered. The high-temperature gas streams produced by combustion can be used in a variety of scenarios for power and heat generation at various scales.

For aluminum, the aluminum-air-based energy conversion system has three stages, the energy storage stage, the energy release stage, and the regeneration stage, as indicated in Figure 6. The energy storage section powers the process of aluminum electrolysis through renewable energy sources [23]. In the energy storage stage, renewable energy is used to power the electrolytic aluminum process. In the energy release process, the aluminum obtained by electrolysis is passed through a combustion power cycle to generate electricity. The chemical energy in aluminum is converted into electricity for transmission to the outside world. The solid-phase products from the combustion of the aluminum powder are also collected. In the regeneration process, the collected solid combustion products are re-electrolyzed into aluminum, which enables the regeneration process of aluminum along with the energy storage process [23]. The aluminum fuel is recycled throughout the process with virtually no consumption. The aluminum-air combustion power generation system is still in the laboratory research period. The research on the aluminum-air combustion mechanism and reaction kinetics, combustor design and development, combustion product morphology and separation, and high-temperature gas heat transfer should be given more attention. Aluminum powder has similar physical properties to solid fossil fuels. If coal-fired power plants are retrofitted for metal fuel combustion, this can significantly reduce infrastructure investment and contribute to zero carbon electricity generation.

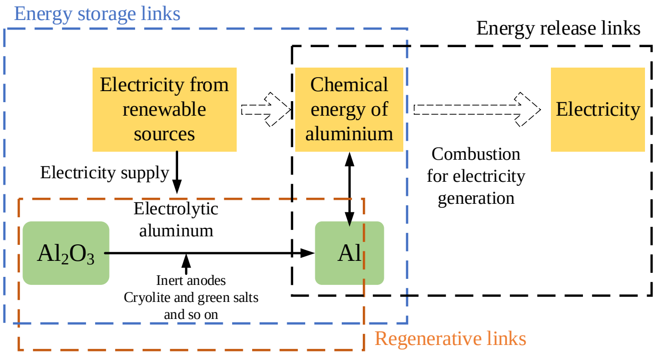


Figure 6. Schematic diagram of the energy conversion principle based on aluminum-air reaction [23].

2.2. Aluminum-Water Reaction Power Generation

The heat generated from the oxidation of metals with water is fully utilized at high temperatures. Otherwise, fuel cells are needed to increase the chemical energy of the hydrogen [12]. The stoichiometric ratio and exotherm of the reaction between aluminum and water vary at various temperatures, and the high-temperature aluminum-water reaction is illustrated in Equation (2).

$$2Al(s) + 3H_2O(g) \rightarrow Al_2O_3(s) + H_2(g) \tag{2}$$

Aluminum-water reaction energy conversion systems are divided into indirect circulation systems and direct circulation systems depending on the reaction cycle [49]. The heat generated by the reaction is coupled to the power generation process by the heat exchanger in indirect-cycle systems, and the reaction process is coupled to the power generation process directly in direct-cycle systems. Indirect-cycle systems approximate coal-fired power generation and direct-cycle systems approximate gas turbine power generation. The principle of the indirect circulation system based on the steam Rankine cycle and the aluminum-water reaction is presented in Figure 7a and the principle of the direct circulation system based on the aluminum-water reaction is displayed in Figure 7b.

(1) Indirect circulation system

As illustrated in Figure 7a, aluminum powder is oxidized by excess water in the aluminum-water reactor to ensure complete and adequate aluminum-water reaction. The heat generated from the reaction is employed to heat the working medium in the steam Rankine cycle. Then, the high-temperature and high-pressure workpiece flows into the steam turbine to do the work and drive the generator to generate electricity. The aluminum-water reaction products include both gas-phase and solid-phase products. The solid-phase products are recovered by collection and then used as feedstock for the alumina electrolysis process in the next energy storage cycle. The gas-phase product is mainly the mixture of high-temperature steam and hydrogen, which passes into the condenser after heat exchange with the circulating working medium in the reactor. The water vapor in the gas-phase mixture is separated from the hydrogen by condensation in the condenser, thus obtaining water and hydrogen. The water separated from the condenser is filtered and then continues to participate in the system cycle. The electricity and hydrogen produced by the system are utilized according to demand.

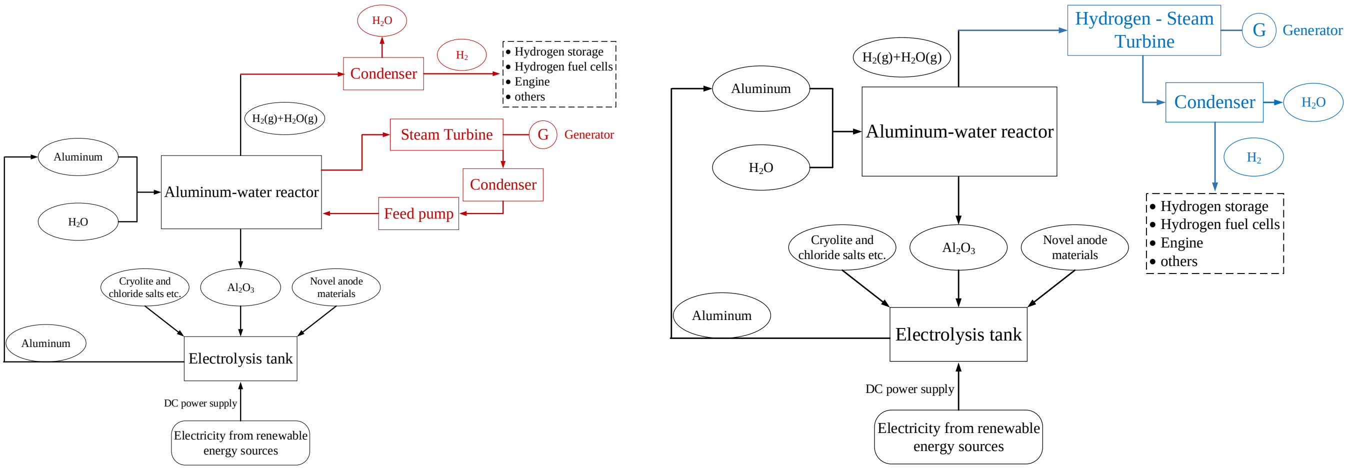
Montorsi et al. [44,60] conducted further research on combustion devices, system construction, and optimization of indirect-cycle systems for aluminum-water reactions. The thermal characteristics of four different circulation systems were analyzed by the combination of aggregated and distributed parameters. The influences of combustion temperature, water-to-aluminum ratio, and combustion pressure on the power generation efficiency, system efficiency, and hydrogen production rate of the cycle system were investigated. It was found that variations in parameters such as combustion temperature, water-to-aluminum ratio, and combustion pressure have effects on the efficiency of the system and have little effect on the hydrogen yield. Yang et al. [61] established an indirect-cycle system based on the aluminum-water reaction and analyzed the influence of key parameters such as cell conversion efficiency through systematic simulations. The system power generation efficiency was 49.25%, improving the battery conversion efficiency, which is beneficial to the utilization efficiency of the system. For aluminum-water-reaction indirect-cycle power generation systems, aluminum-water reaction kinetics, aluminum-water reactor design, and system design all require further research.

(2) Direct circulation system

As indicated in Figure 7b, the high-temperature and high-pressure gas mixture from the aluminum-water reaction in the direct-cycle system flows directly into the hydrogen-steam turbine to do the work and drive the generator to produce electricity. Compared to the aluminum-water reactor in an indirect circulation system, the aluminum-water reactor in a direct circulation system has to be at high temperature and pressure, which places higher demands on the equipment. The aluminum-water reactor is the critical equipment in the direct circulation system. Turbines and fuel cells are also used to harness the mixture of gases generated by the aluminum-water reaction [1,12,62].

The internal combustion engine enables clean and efficient utilization of the high-temperature and -pressure hydrogen from the reaction. Interception of the solid-phase products after the reaction is required in order to avoid wearing out the generator. In addition to internal combustion engines, external combustion engines are also available for direct circulation systems, a system in which a hydrogen-steam mixture is combusted together with air with high efficiency [1]. Research on aluminum-water combustion direct-cycle systems has focused on aluminum-water reaction characteristics at high pressure, aluminum fuel delivery devices, cycle system construction, and system thermal characteristics.

Vlaskin et al. [62] constructed an experimental hydrogen cogeneration plant based on an aluminum-water reaction. The average reaction temperature and pressure of the system were 324 °C and 15 MPa, respectively. The electrical efficiency of the power plant was 12% and the total efficiency was 72%. Vlaskin et al. [63] also found that the efficiency of power plants was improved by utilizing the mixture of gases from the aluminum-water reaction via a gas turbine. Based on the aluminum-water reaction, Yang et al. [61] constructed the direct-cycle co-generation system. The system produced approximately $22.2\ \mathrm{MJ}\cdot\mathrm{kg}_{(\mathrm{Al})}^{-1}$ of heat and electricity with a power generation efficiency of 41.52%. Franzoni et al. [12] calculated the energy conversion efficiency of the aluminum-water reaction system when superheated steam cycles and combined heat and power cycles were used. Hydrogen from the aluminum-water reaction for fuel cells had system efficiencies in the range of 0.62–0.85. In contrast to aluminum-air energy conversion systems, aluminum-water reaction energy conversion systems produce and utilize hydrogen. The increasing complexity of the system has the potential to reduce its efficiency [37]. The aluminum regeneration process is the critical link for aluminum as a recyclable energy carrier. Aluminum electrolysis is a high-energy-consuming industry, and the electricity consumption in the aluminum regeneration process is significantly influencing the economy of the aluminum-based fuel energy conversion system. If the electricity generated by new energy is utilized for aluminum regeneration, not only is the volatility of new energy generation alleviated, but the cost of electricity consumption of the system is also reduced. The economy of the system is improved. For the aluminum-water reaction direct circulation system, the relevant work is still mainly in the basic research of aluminum-water high-pressure combustion in the laboratory, the construction and optimization of the circulation system, and the analysis of the system economy [12]. Aluminum-water high-pressure combustion reaction kinetics, aluminum-water combustion reactor development, the system key equipment design, and large-scale system integration still need further in-depth research to achieve engineering applications.



(**a**) (**b**)

Figure 7. Schematic diagram of the energy conversion principle based on the aluminum-water reaction [64]. (**a**) Indirect circulatory system. (**b**) Direct circulation system.

3. Combustion Characteristics of Aluminum

The aluminum powder has to be combusted at high temperatures and pressures to obtain high energy and power density. If only hydrogen is required, aluminum powder can be reacted with water at low to medium temperatures. Relevant research examines aluminum as fuel reaction characteristics with different application scenarios.

3.1. Aluminum-Air Combustion Characteristics

Aluminum has a high reactivity and can be oxidized at room temperature [65]. The oxidized particles are covered by an amorphous oxide layer. The oxide layer prevents further oxidation of the aluminum inside [66]. At lower heating rates, increasing the temperature promotes interdiffusion between Al^{3+} and O^{2-}. The aluminum particles are oxidized layer by layer from the inside out. The reaction lasts for a long time and is also known as the dynamic (non-isothermal) oxidation process of aluminum powders. Figure 8 illustrates the oxidation mechanism of aluminum particles at room temperature. Caberra-Mott proposed the mechanism of room-temperature oxidation of aluminum [64]. The mechanism assumes that the initial oxide layer on the aluminum surface separates the active aluminum inside from the oxygen. Due to the migration of electrons, Al^{3+} and O^{2-} accumulate on the inner and outer surfaces of the oxide layer, respectively. A uniform electric field is formed within the oxide layer. Driven by an electric field, Al^{3+} crosses the oxide layer and reacts with the oxygen in the environment to form new Al_2O_3. The oxide film grows and thickens gradually, as depicted in Figure 8. The oxidation rate is determined by Al^{3+} and the electron migration rate [67].

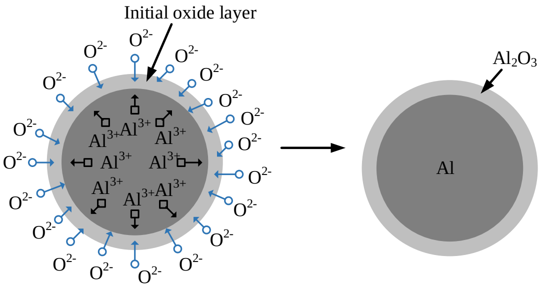


Figure 8. Schematic diagram of aluminum oxidation mechanism at room temperature [64].

Studies on the combustion characteristics, combustion mechanism, ignition temperature, and agglomeration characteristics of solid products of aluminum powder are constantly being enriched and refined [68]. Mikhaylo et al. [69] analyzed the oxidation products of micron aluminum powder when heated to various temperatures in oxygen atmosphere. It was found that the oxidation products were in the order of amorphous alumina layer $\rightarrow \gamma\text{-}Al_2O_3 \rightarrow \delta\text{-}Al_2O_3 \rightarrow \theta\text{-}Al_2O_3 \rightarrow \alpha\text{-}Al_2O_3$ as the temperature increased. The ignition temperature is a critical parameter in the investigation of the ignition and combustion characteristics of aluminum powders and usually varies over a wide range as the reaction conditions alter. Factors such as the type and content of oxidant in the environment and pressure also influence the ignition and subsequent combustion of aluminum powder. Bazyn et al. [70] investigated the combustion characteristics of 10 μm aluminum powder under the oxygen atmosphere and found that the combustion time of the powder reduced as the pressure escalated. This is primarily because the combustion of aluminum powder in O_2 is a reaction-kinetics-controlled process, whereas its combustion in H_2O and CO_2 is a diffusion-controlled process [71].

According to various research methods, the theoretical models of the aluminum powder combustion mechanism can be divided into the following three categories. (1) Semi-empirical model. Single-particle combustion is chiefly studied by assumptions and simplified calculations, but, in practice, the combustion is mainly in groups of particles. (2) Kinetic mechanism model of chemical reaction [59]. The evolution of the reaction paths during the reaction is mostly evaluated by reaction kinetics software such as Chemkin. (3) Computational fluid model, namely the CFD model [72]. It is mainly used to simulate flame propagation, pressure fields, temperature fields, and concentration fields during combustion. The D_n model is a typical semi-empirical model and provides the basis for the further development of combustion mechanism models. Law built the Law model based on the D_n model and the mass–energy conservation equation, as presented in Figure 9 [73]. Thereafter, Liang and Beckstead developed a two-dimensional, non-stationary Liang–Beckstead model based on the Law model, taking into account thermal radiation, convection, and cohesive phase oxidation product aggregation (oxidation caps), as illustrated in Figure 10 [74]. Both the Liang–Beckstead model and the Law model are chemical reaction mechanism models that have been extensively used in subsequent studies. All the above mechanistic models are single-particle models with aluminum powder. The CFD model allows the two-phase flow of the particle population with the oxidizer to be studied, which is more instructive for practical engineering. The CFD models are in the process of developing.

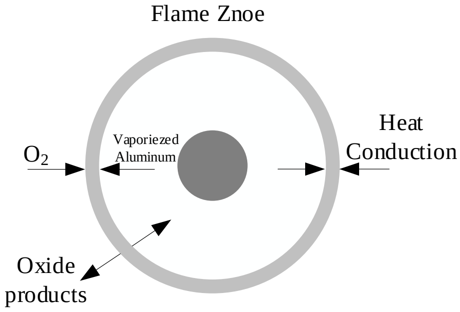


Figure 9. Aluminum particle combustion model of Law [73].

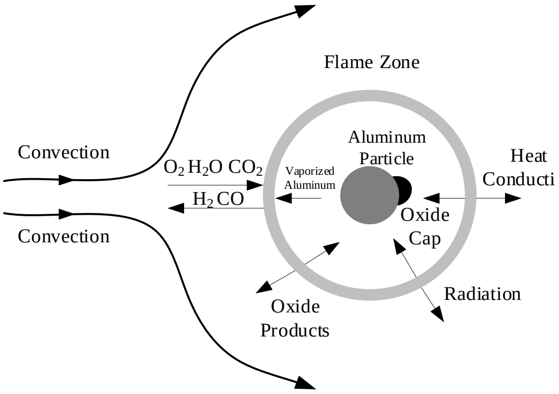


Figure 10. Aluminum combustion model for Liang–Beckstead [74].

The combustion process of aluminum powder produces cohesive phase products, of which agglomerates are essential for combustion. The research on aluminum combustion agglomeration has focused on agglomeration mechanisms, aluminum combustion, and condensed phase products. Pressure, combustion rate, and aluminum powder particle size all have implications for agglomeration. The influence of the various factors on agglomeration depends essentially on the competing processes of molten aluminum powder agglomeration and aluminum agglomerate ignition. Maggi et al. [75] observed the agglomeration process of aluminum particles during combustion through experiments, and the combustion behavior was consistent with the Liang–Beckstead model. Further research is still needed on the prediction and inhibition of agglomeration during the combustion of aluminum powders and the mechanism of agglomeration under high pressure.

3.2. Aluminum-Water Combustion Characteristics

The presence of Al_2O_3 on the aluminum surface causes aluminum to be almost impervious to reaction with water at room temperature [76–78]. However, aluminum is gradually oxidized by water as the temperature rises [79]. Depending on the reaction temperature, the aluminum-water combustion reaction and the conversion of the reaction products are described in Table 1 [11,80]. The temperature range varies with the specific experimental conditions and the temperatures in Table 1 are only a reference range. The same mass of aluminum powder produces the same amount of hydrogen, but the stoichiometric ratio of the reactants changes as the temperature increases. Both $Al(OH)_3$ and $AlO(OH)$ transform to Al_2O_3 as the temperature increases [80]. Huang et al. [81] believed that the formation rate of Al_2O_3 was slow due to the lack of oxygen atoms in the aluminum-water reaction. Ngenito et al. [82] divided the reaction of liquid aluminum with water into two parts: the surface reaction and the gas-phase reaction. Some studies have also analyzed the mechanism of the aluminum-water reaction from the molecular perspective through simulations [83]. Russo et al. [84] believed that the oxidation layer on the aluminum surface reduced the reactive site of aluminum, thus preventing the aluminum-water reaction at room temperature. Roach et al. [85] considered from experimental studies that the reactivity of aluminum in reaction with water is affected mainly by the geometric configuration of aluminum.

According to previous studies [77,86,87], the low-temperature aluminum-water reaction has three stages, as depicted in Figure 11. There is a layer of amorphous alumina on the surface of aluminum powder. A weak and rapidly quenched initial oxidation stage occurs when the aluminum powder meets water at first. The amorphous alumina initiates hydrolysis during the induction phase. Once the oxide layer has been sufficiently destroyed, hydrogen and heat are rapidly released, until the tough hydroxide is formed and the reaction is inhibited. The reaction may leave unreacted aluminum nuclei, resulting in low yields. The three pathways of the aluminum-water reaction are given in Table 1. Despite the different stoichiometry of water, the ratio of aluminum to the hydrogen produced is invariably the same. Shrinking core models are often used for kinetic analysis of aluminum-water reactions [88–90]. However, the shrinking core model is only for spheres of constant size and with constant diffusion rates [91–95]. Razavi-Tousi et al. [77] and Wang et al. [96] improved the shrinking core model for hydrogen production by aluminum-water reaction. The initial phase in the reaction is controlled by the diffusion of water molecules through the product layer. The initial stage has a loose product layer with a slight thickness. The transport of water molecules meets the reaction. This stage is controlled by the surface chemistry [97]. The oxide layer gradually accumulates on the aluminum powder surface as the reaction continues. The transport of water molecules to the surface of the unreacted aluminum particles is blocked. The water molecules passing through the reaction layer become the control step at this point. The aluminum-water reaction becomes more difficult to carry out. It is necessary to further activate the reaction so that the unreacted aluminum nuclei can participate in the reaction and improve the reaction efficiency. Yavor et al. [97] investigated the reaction of nano- and micron-sized aluminum powders with water at various temperatures and proposed an expression for predicting the maximum hydrogen production rate at various powder sizes and temperatures. It was found that the aluminum-water reaction was inhibited after reaching the penetration thickness of the aluminum particles, which was temperature-dependent and independent of particle size. Some researchers have also increased the temperature and pressure of water to supercritical to react with aluminum, which is strongly oxidized by supercritical water [98].

Table 1. Aluminum-water reactions at various temperatures [11,80].

Temperature Ranges	Aluminum-Water Reaction Equation	Reaction Product Conversion
<200–300 °C	$2Al + 6H_2O \rightarrow 2Al(OH)_3 \downarrow +3H_2 \uparrow (3)$	$Al(OH)_3 \rightarrow AlO(OH) + H_2O \uparrow (6)$
~200–570 °C	$2Al + 4H_2O \rightarrow 2AlO(OH) \downarrow +3H_2 \uparrow (4)$	$2AlO(OH) \rightarrow Al_2O_3 + H_2O \uparrow (7)$
>570 °C	$2Al + 3H_2O \rightarrow Al_2O_3 \downarrow +3H_2 \uparrow (5)$	$Al(OH)_3 \rightarrow Al_2O_3 + 3H_2O \uparrow (8)$

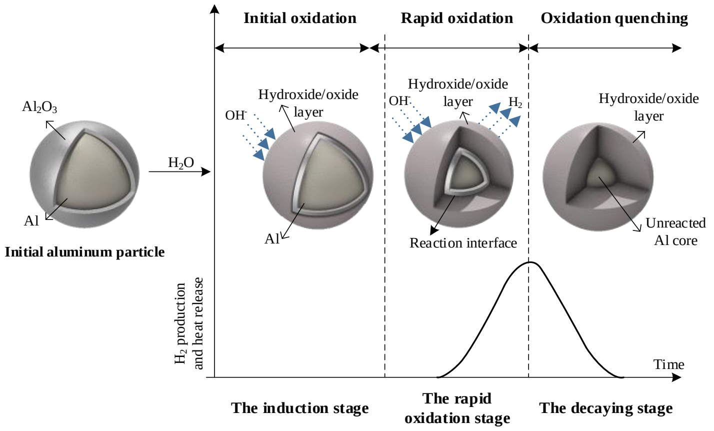


Figure 11. Schematic diagram of low-temperature aluminum-water reaction mechanism [77,86,87].

The passivation layer on the aluminum powder surface is the main obstacle to the fast and efficient aluminum-water reaction [99]. The activation of the aluminum reaction revolves around the destruction of the passivation layer and the inhibition of its production [7]. The current activation methods include alkaline activation, alloying activation, additive activation, mechanical activation, and high-temperature aluminum-water reactions.

(1) Activation of alkaline or acidic solutions

Aluminum is a typical amphoteric metal that reacts with both acids and alkalis [100]. It was found that aluminum reacts better with alkalis. Activation of the aluminum-water reaction is mainly by alkalis. The essence of alkali activation is electrochemical corrosion, as shown in Equations (9)–(12) [7,11,33,99,101–103].

$$Al_2O_3 + 3H_2O + 2OH^- \rightarrow 2Al(OH)_4{}^- \tag{9}$$

$$Al + 4OH^- \rightarrow Al(OH)_4{}^- + 3e^- \tag{10}$$

$$2H_2O + 2e^- \rightarrow H_2 \uparrow + 2OH^- \tag{11}$$

$$Al(OH)_4{}^- \rightleftharpoons Al(OH)_3 \downarrow + OH^- \tag{12}$$

The reversible reaction (Equation (12)) occurs in the presence of an excess of $Al(OH)^{4-}$, which prevents the reaction from continuing. A high concentration of alkali is required to maintain the reaction, usually requiring more than 10% by mass of NaOH. The superposition of Equations (8)–(10) results in Equation (1). The alkali is eliminated during the reaction. The alkali can be seen as the “catalyst” for the reaction, eliminating the passivation layer while preventing the aluminum surface from passivating again. NaOH, KOH, $Al(OH)_3$, and $Ca(OH)_2$ have been widely investigated and applied with high activation results [7,33,45,78,101,102,104–111]. To reduce the corrosion of equipment, alkaline solutions such as $NaAlO_2$, Na_2SnO_3, and KF were also found to have a significant activating effect [88]. The alkali activation allows the hydrogen production rate of the aluminum-water reaction to be increased to 100% at room temperature with the short reaction time. The alkaline activation is simple to operate and can be reacted efficiently at room temperature with low cost. The high corrosiveness of the alkaline solution places higher demands on the equipment. In addition to improving the resistance of the material, the combination of other activation methods to achieve a good activation effect even at low concentrations of lye is worth further investigation [102,107,110].

(2) Activation of alloys

The alloys are primarily prepared by smelting and ball milling [11]. The major activation principles for alloying are the primary cell and eutectic effects [109,112]. The alloy prepared by ball milling forms defects on the surface during the ball milling process. The increased number of grain boundaries also contributes to the reactivity [11,76,113]. Low-melting-point metals are usually added to aluminum to form alloys [114]. Common low-melting-point metals include Li, Ga, In, Sn, and Bi [11,19,44,107,112–119]. The alloy has a high activation effect [113,120,121]. The addition of cheaper metals for activation of the aluminum-water reaction to reduce the cost of alloying, such as Cu, Mg, and Fe, has also been investigated [109,113,119,121]. The alloying has achieved high results and has the potential for mass production. Nevertheless, the cost of alloy preparation is relatively high and the preparation process is potentially contaminated. The alloy produced by ball milling also suffers from storage difficulties and high electricity consumption [76]. In addition to the development of low-cost aluminum-based alloys, the safety of the alloy preparation process has to be improved and effective storage methods developed.

(3) Activation of additives

The addition of various activators by ball milling destroys the oxide film on the aluminum particles surface and allows the aluminum to react with neutral water at room temperature. The physicochemical properties of the additive itself influence the reactivity of the aluminum. Pitting is one of the principal causes of additive activation. Oxide additives, such as AlO(OH), $Al(OH)_3$, and Al_2O_3 [86], and water-soluble inorganic salt additives, such as NaCl, KCl, and $AlCl_3$, are popular activators for aluminum-water reactions [87,102,108,122–126]. The additive develops pitting during ball milling, destroys the passivation layer on the aluminum surface, and inhibits the agglomeration of aluminum during ball milling. Some studies have been conducted to improve aluminum-water reactivity by adding carbon materials such as olefin carbon, graphite, and silicon carbide [127]. Hydrides are frequently used for hydrogen storage and react with water violently, making the solution alkaline and releasing large amounts of hydrogen and heat [17]. It is also utilized for aluminum-water reaction activation such as $NaBH_4$, $LiBH_4$, LiH, and CaH_2. Adding additives to aluminum effectively reduces the reaction conditions. However, the response time becomes longer and continuous reaction proves difficult. The preparation and storage requirements of the composites are high and some of the additives are expensive. Additives also decrease the hydrogen storage density.

(4) Mechanical activation

The aluminum-water reaction can be activated by mechanical means only, such as cutting and grinding [128]. Mechanical activation can take place both before and during the reaction. When mechanical activation occurs during the reaction, activation and oxide formation take place simultaneously. The newborn aluminum is then oxidized due to pitting. The method of producing hydrogen while wet-cutting aluminum alloys is known as mechanical activation in the reaction [129]. Without activation, the conversion of industrial micron aluminum powder is far below 100% below 100 °C, even after many hours of full agitation [130,131]. Nano-aluminum powders have high chemical activity [59]. Ivanov et al. [130] and Schoenitz et al. [131] found that the rate of hydrogen production and the hydrogen yield in a short period of time were higher, close to 100%, compared to micron aluminum powder under similar physical environment conditions without other activation methods. The aluminum nano-powder enhances the reaction effect to some extent. Compared to micron aluminum powders, nano-aluminum powders have decreased active aluminum content and apparent density [27]. It is difficult to store and transport aluminum nano-powders and there are possible safety issues. Nanometer aluminum powder has high preparation cost and is easy to be oxidized. It is difficult to apply at scale.

(5) High-temperature activation

The aluminum-water reaction can be activated by the high-temperature environment [132]. Micron aluminum powders are available for more applications at higher temperatures. Vlaskin et al. [79] investigated the kinetics of oxidation of micron aluminum powder in high-temperature boiling water through experiments. It was found that micron-sized aluminum powder can be intensely oxidized in boiling water above 230 °C for a short period of time, with a hydrogen conversion rate close to 100%. Trowell et al. [98] performed the reaction of both micron-sized aluminum powder and crude aluminum (including aluminum lumps, sheets, and aluminum cans) with water without any catalyst at temperatures ranging from 475 K to 650 K (including the corresponding saturation vapor pressure of the water). Below 550 K, the hydrogen yield of aluminum powders with a particle size greater than 50 μm was less than 30% and the crude aluminum hardly reacted with water. The hydrogen yield of all samples was greatly improved when the temperature was increased to 550 K to supercritical temperature. Hydrogen production reached 100% for all samples at temperatures above 647 K and pressures above 22 MPa (the critical point for water). It was demonstrated that 100% hydrogen production was also achieved by increasing the reaction temperature and pressure of crude aluminum with water. The high temperature provides the higher thermodynamic efficiency [133]. The high-temperature aluminum-water reaction provides novel insights into circulating power systems. Nevertheless, the high-temperature aluminum-water reaction demands high initiation temperatures and the application scenarios are limited.

Table 2 contains the comparison of the properties of various activation methods for the aluminum-water reaction. The degree of reaction and reaction rate of the aluminum-water reaction, the production–storage–transport of the reactants, and the recovery of the reaction products are all factors to be considered in practical applications. The different application scenarios place various demands on the response.

Table 2. Comparison of the properties of different activation methods for the aluminum-water reaction.

Activation Method			Key Activation Mechanisms	Application Areas
Changing the environment	Acidic or alkaline solutions (Overwhelmingly alkaline solutions)		Electrochemical corrosion	Portable Transport
	High-temperature aluminum-water reaction		Melting of the passivation layer	Stationary
	Mechanical activation of aluminum (Mainly aluminum nano-powder)		pitting corrosion	Portable
Changing the aluminum properties	Aluminum-based alloys	High-temperature melting	Principle of Primary Battery Eutectic reaction	Portable
		Mechanical ball milling	Principle of Primary Battery Eutectic reaction Pitting corrosion	Portable
	Non-metallic additives	Oxidation	pitting corrosion Principle of Primary Battery	Portable
		Inorganic salts	pitting corrosion Principle of Primary Battery	
		Cyanides	Principle of Primary Battery	
		Carbon material	Nuclear pore structure	

4. Aluminum Regeneration

Aluminum has a proven regeneration process. High-purity aluminum metal is obtained by refining aluminum-containing scrap and electrolytic alumina [27,134]. The aluminum obtained from the melting of aluminum scrap and aluminum scrap is called recycled aluminum and is mainly in the form of aluminum alloys [135]. Recycled aluminum is capable of recycling aluminum and consumes less electricity [136]. The recycled aluminum is mainly smelted by fire [137]. The use of fossil fuels in the smelting process also results in the burning of large amounts of pollutants, which pollute the environment [138]. Consequently, this work focuses on the process of aluminum regeneration through electrolysis. Bai et al. [49] found that the energy storage efficiency of the electrolytic aluminum had a significant impact on the round-trip efficiency of the system. Further reduction in the electricity consumption of electrolytic aluminum is also the focus of subsequent research.

Aluminum in the geological crust exists mainly in the form of aluminum silicate. Crystalline alumina–alumina (Al_2O_3) is the main raw material to produce aluminum and most metallurgical uses of alumina are bauxite [27]. Bauxite is dominated by alumina trihydrate ($Al(OH)_3$) and alumina monohydrate ($AlO(OH)$), together with various compounds. Currently, alumina is often extracted from bauxite by the Bayer method [139]. The solid products of the aluminum-water reaction at different temperatures are mainly Al_2O_3, $AlO(OH)$, and $Al(OH)_3$. The solid-phase product of the combustion of aluminum in air is Al_2O_3. Regeneration of aluminum can be achieved by re-electrolysis of the collected solid products of aluminum combustion. At present, the process of electrolytic aluminum is well established in the industry. Cryolite (Na_3AlF_6) is usually used as the electrolyte [27]. High-purity aluminum is obtained by electrolysis of a combination of cryolite and molten alumina. In theory, the aluminum electrolysis process does not lose cryolite, but in practice, it is consumed in large quantities. The entire aluminum electrolysis process is illustrated in Equation (13).

$$Al_2O_3 \rightarrow 2Al + 1.5O_2 \tag{13}$$

Electrolytic technology for carbon anodes is used in industrial processes (Hall–Héroult process) [140]. The process of aluminum regeneration produces CO and CO_2. Carbon-anode-based electrolytic aluminum technology melts metallurgical-grade alumina in a high-temperature cryolite bath for electrolysis. The carbon anode consists mainly of calcined coke and bitumen as a binder. Two types of anode are presently used: self-baking and pre-baking. The reaction equation is shown in Equation (14).

$$Al_2O_3{+}nC \rightarrow 2Al + (3-n)CO_2{+}(2n-3)CO \tag{14}$$

The Hall–Héroult process produces aluminum with a purity higher than 99%, but the carbon anode is constantly oxidized during the electrolysis process. Including emissions from power plants, each 1 kg of aluminum produced by electrolysis releases 8 kg of CO_2 equivalent [141]. While many greenhouse gases are discharged, solid waste such as sulfur, fluorine, and anodic carbon residue will also pollute the environment [142]. The adoption of inert anodes makes aluminum production more environmentally friendly, but also adds more electrical input. Aluminum electrolysis itself requires a large amount of electricity [142], with one ton of aluminum electrolysis consuming around 13,000 kWh. The cost of electricity consumption is also one of the issues that needs to be addressed. Aluminum electrolysis has always been recognized as a high-energy-consuming and -polluting industry [142]. The development of zero-carbon energy-saving aluminum recycling technology is also an effective way to alleviate the contradiction between resources, energy, and environmental management [141]. According to Faraday's law, the electrical energy consumed by electrolytic aluminum is determined by the average voltage and current efficiency, as shown in Equation (15).

$$Al_2O_3 + nC \rightarrow 2Al + (3{-}\,n)CO_2{+}(2\,n\,{-}3)CO \tag{15}$$

where w is the electrical consumption, kWh-t_{Al}; γ is the current efficiency, %; $E_{average}$ is the average voltage.

According to Equation (1), the energy saving of electrolytic aluminum should be carried out in two directions: reducing voltage and improving current efficiency. Aluminum fuel regeneration is the significant aspect to determine the economy and feasibility of energy storage systems based on aluminum fuel. There is still plenty of scope for research and development in the area of efficiency and emissions reduction in aluminum electrolysis technology. The uses of renewable electricity, inert anode material technology, and wettable cathode technology are all effective in improving the energy efficiency of the aluminum electrolysis process and reducing carbon emissions.

(1) Inert anodes

When inert anode materials are employed in the aluminum electrolysis process, the anode product is O_2 and no more carbon emissions are produced [143]. With inert anodes, the anode is not consumed and there is essentially no necessity to replace the anode material. The electrolysis tank is available in a completely enclosed design, reducing the emission of harmful gases and particulate pollutants. The inert anode is expected to achieve zero emissions in the electrolysis process. Inert anode materials require good electrical conductivity and resistance to thermal shock, as well as the ability to withstand erosion by molten fluoride and percolation of neo-ecological oxygen above 1000 °C. The research in inert anode technology includes metals, alloys, composites, oxides, and ceramics [143–145]. The metal-ceramic inert anode has the characteristics of both metal and ceramic. The corrosion resistance, electrical conductivity, and mechanical properties of metal-ceramic inert anodes are decisively influenced by their preparation process. $NiFe_2O_4$-based metal-ceramic inert anodes have been widely concerned [146]. The $NiFe_2O_4$-based cermet inert anodes were modified by the addition of NiO, Cu, and Nb_2O_5, with the research focusing on thermal shock resistance and electrical conductivity [147]. The introduction of metals was also found to increase the thermal conductivity and reduce the thermal difference of cermet, and the addition of nickel improved the material properties of $NiFe_2O_4$-based materials [148,149].

(2) Wettable cathode technology

The molten aluminum wets well with the wettable cathode surface [150]. The use of wettable cathode technology in aluminum electrolysis creates a flat and stable cathode by simply hanging a 3–5 mm film of aluminum solution on the anode. The anode is not required to preserve a layer of molten aluminum of about 20 cm. The interference of the magnetic field with the electrolysis process is eliminated, the distance between two stages is shortened, and the energy consumption is reduced. The electrolytic efficiency is improved. TiB_2 wettable cathodes have high wettability with molten aluminum and have potential as a material for cathodes in Hall–Héroult process electrolyzers [151]. The current problem of TiB_2 wettable cathodes focuses on improving the sintering characteristics and the mechanical properties of TiB_2 materials, and reducing corrosion rates, resistivity, and costs while extending material life [151,152]. The optimum wetting angle between the TiB_2 wettable cathode and the molten aluminum has been impossible to determine [150]. The wetting sequence of aluminum and salt is difficult to resolve [153]. These issues have been extensively and intensively researched with some results. However, it is still a long way from large-scale industrial applications.

5. Summary and Outlook

Environmental problems prompt countries to promote the transformation of the energy structure. The active development of large-scale and long-cycle energy storage technologies will also contribute to meet the demand for low-carbon energy, alongside the vigorous development of renewable energy. Although energy storage technology has come a long way, there has never been a universal storage technology. Each energy storage technology has its own dedicated area. There is no proven energy storage technology that can meet the needs of global trade.

Hydrogen is considered a potential zero-carbon energy source, and its production, storage, transportation, and application technology have always been the focus and difficulty of research. As a typical clean, recyclable energy carrier, metal fuels are expected to play a significant role in low-carbon energy. Among them, aluminum has a high energy density and low carbon pollution. Aluminum can react with water to produce hydrogen and its combustion products have a proven recycling process. Aluminum is also globally traded and is considered to be a promising carrier for energy and hydrogen storage.

Aluminum not only burns in the air but also reacts with water to produce hydrogen while releasing large amounts of heat. In addition to batteries, the aluminum combustion power cycle system is also employed to convert the chemical energy of aluminum fuel into electrical energy to achieve energy release and conversion. The heat generated by the reaction is fully utilized. The solid products of aluminum combustion are transported, and then the conversion from electricity to chemistry and storage can be achieved through the electrolysis of aluminum oxide. Energy is stored and recycled in the process. Energy conversion systems based on aluminum fuel storage are environmentally friendly and have high energy density. The system has a long energy storage cycle and the fuel is recyclable. In situ hydrogen production is also achieved based on the aluminum-water reaction.

However, the technology is still some way from being industrialized on a large scale. The design and development of metallic aluminum reactors and the associated heat transfer calculations are key to the realization of energy conversion systems. The dense oxide layers on the aluminum surface are an important obstacle to the reaction. In order to promote the realization of fuel aluminum energy storage technology, the subsequent research on aluminum reaction kinetics should be continued, including aluminum-air reaction kinetics and aluminum-water reaction kinetics. The construction and optimal design of energy conversion systems based on aluminum-based fuel are also a focus of subsequent research. The energy storage efficiency of electrolytic aluminum has an important influence on the whole energy conversion system. The clean and efficient electrolytic aluminum technology should also be the focus of research.

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