



A comprehensive review of carbon and hydrocarbon assisted water electrolysis for hydrogen production

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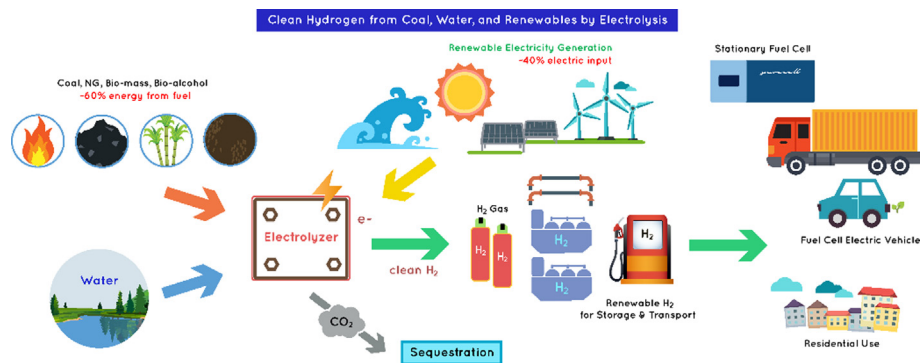
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HIGHLIGHTS

- Carbon/hydrocarbon assisted water electrolysis reviewed for clean hydrogen production.
- Reviewed different electrochemical technologies using carbon fuels under development.
- This technology has been progressed to different levels of maturity for different carbon sources.
- This route of hydrogen production can lower the electric input and CO₂ emissions from the carbon sources.

GRAPHICAL ABSTRACT



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ABSTRACT

Hydrogen is mainly produced by natural gas reforming, which is a highly efficient process with low feedstock costs. However, the rising interest in clean technologies will increase the demand for hydrogen, meaning that other sources will need to be explored. Although coal is currently the major source of power generation, its demand appears to be declining due to the rise in electricity generated from renewable energy sources and the worldwide quest for low-emission power generation. Coal reserves worldwide are abundant, but new technologies would be needed to produce hydrogen from this feedstock. Coal gasification is one well-established technology for this purpose, but it is inefficient and produces high CO₂ emissions. An alternative technology that has been investigated over the past few decades is carbon assisted water electrolysis. The basic process is water/steam electrolysis, with part of the energy required for the electrolysis provided by the chemical energy of coal,

Abbreviations: ACFC, air-carbon fuel cell; AEM, anion exchange membranes; CAWE, carbon/coal assisted water electrolysis and carbon/hydrocarbon assisted water electrolysis; CHP, combined heat and power; COD, chemical oxygen demand; COR, carbon-water oxidation reaction; DCFC, direct carbon/coal fuel cells; DHA, 1,3-dihydroxyacetone; DMSO, dimethylsulphoxide; EAWE, ethanol assisted water electrolysis; FE, Faradaic efficiency or coulombic current efficiency; GC, gas chromatography; GDC, gadolinium-doped ceria; GDL, gas diffusion layer; HER, hydrogen evolution reaction; HHV, higher heating value, liquid water; HMF, hydroxymethylfurfural; HOR, hydrogen oxidation reaction; LHV, low heating value, steam water; LLNL, Lawrence Livermore National Laboratory; LMA, liquid metal anode; LSM, Sr-doped lanthanum manganite; MAWE, methanol assisted water electrolysis; MEA, membrane electrode assembly; NDIR, non-dispersive infrared detector; NG, natural gas; OCV, open circuit voltage; OER, oxygen evolution reaction; ORR, oxygen reduction reaction; PEM, proton conducting membrane and proton exchange membrane; PEMFC, polymer electrolyte membrane fuel cell; PSEM, proton-conducting solid electrolyte membrane; PTFE, polytetrafluoroethylene; SCAFC, steam-carbon-air fuel cell; SCFC, steam-carbon fuel cell; SMR, steam methane reforming; SOE, solid oxide electrolyte; SOEC, solid oxide electrolysis cell; SOFC, solid oxide fuel cell; SOM, small organic molecules; SPEEK, sulfonated polyetheretherketone; TOC, total organic carbon; WGS, water gas shift; YSZ, yttria stabilized zirconia

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which reduces the overall electrical energy input. In addition to coal, the process can also use other carbon sources, such as biomass, alcohols or gaseous hydrocarbons. Several studies have investigated this electrochemical route of hydrogen production, employing different electrolytes in a wide temperature range (room temperature to 850 °C) under different process conditions. This paper presents a comprehensive review of carbon assisted water electrolysis, associated materials used and the challenges for the development of the technology at the commercial scale.

1. Introduction

Solutions for global energy challenges are being sought by moving towards eco-friendly technologies for a sustainable future. In an effort to reduce dependence on fossil fuel based energy sources, there has been substantial activity using renewable energy sources (e.g. solar, wind, hydro, ocean and geothermal). However, many such energy sources require substantial initial investment, and the cost of electricity generation can be high. Moreover, the intermittency of renewable energy sources and their dependence on environmental conditions places a strong emphasis on the need for cost-effective, large-scale storage solutions.

Hydrogen is an excellent energy storage medium, the use of which in fuel cells for stationary, transport and portable power applications offers several advantages. These include high efficiency, low greenhouse and pollutant (NO_x, SO_x, and particulates) emissions at the point of use, economies of scale (being modular in nature), and distributed and combined heat and power (CHP) generation. The majority of hydrogen produced at present (about 60 million tonnes per annum) is used for ammonia production, in oil refineries to generate a lighter oil fraction, and to a smaller extent in the chemical and pharmaceutical industries, space exploration and power generation. Most of this hydrogen is produced either by natural gas (NG) reforming with an efficiency of around 70–75%, or by coal gasification, which is about 45–65% efficient [1–5] and has high CO₂ emissions. Generally, these processes are suitable for large scale hydrogen production in central plants, which also offer a favourable cost structure. Only a small percentage of worldwide hydrogen production is via water electrolysis. Although suitable for distributed hydrogen production at demand centres, the process is very energy intensive, requiring 6–7 kWh electric input per m³ of hydrogen produced (65–80 kWh kg⁻¹ at 25 °C and 1 atm), and the capital cost and the cost of production are very high [6–8].

Coal is currently used to meet around 40% of total global electricity demand and is forecast to remain a major source for power generation for many more decades, due to its low cost and vast reserves [9]. However, due to the low efficiency of its conversion to electricity (typically 35–40%), it can contribute significantly to the CO₂ and other pollutant emissions. Although carbon capture and storage (CCS) technologies have the potential to reduce CO₂ emissions to the atmosphere, their large-scale deployment to capture even a small percentage of the 32 billion tons per annum of CO₂ generated globally and the further power generation efficiency losses resulting from this process means a range of solutions are required.

The electrochemical oxidation of solid carbon has been under investigation for many decades for power generation; for example, in direct carbon/coal fuel cells (DCFC) for efficient conversion of coal to low-emission electricity [10–16]. New electrochemical technologies are also currently under development that combine water electrolysis with the use of coal or hydrocarbon fuels to assist with the electrochemical reactions and reduce the overall energy consumption [17–21]. Thermodynamic and operational analysis of these systems suggest that about 60% of the energy required for water electrolysis can be provided by coal or other hydrocarbon (methanol, ethanol, NG) fuels in the form of chemical energy, thus substantially reducing the electric input [20–28]. Furthermore, if the process can be coupled with a renewable energy source, the intensity of CO₂ emissions can be significantly

reduced and hydrogen can be generated at distributed or end-use sites as required by emerging power-generation technologies, such as fuel cells.

Carbon/hydrocarbon assisted co-electrolysis has the potential to offer a less energy-intensive and relatively clean method of using the chemical energy in various sources of carbon, such as low-rank coals, alcohols, methane, NG and even biomass, where pure CO₂ produced can be easily sequestered without the need for separation. In this article, we comprehensively review different low and high temperature electrochemical technologies currently under development, which use a range of electrode and electrolyte materials such as aqueous, polymer electrolyte membrane and solid oxide electrolyte (SOE) membrane cells. The review also covers:

- Various types of carbon sources (coal, methanol, ethanol, glycerol, ethylene glycol, cellulose, NG, methane and CO) used to assist the water electrolysis process
- The electrochemical reactions involved and electrode kinetic limitations
- Thermodynamic properties, such as free energy and enthalpy change of reactions and theoretical cell voltages required
- The energy input required for the electrolysis process with different types of carbon sources and under different process conditions

We also summarise and discuss the results obtained by various investigators in terms of cell designs, materials used, current densities obtained at given voltages, and electric energy savings in the electrolysis process. The review concludes with a discussion of the outlook for technical challenges and new opportunities in this field.

2. Fuel sources to assist with water electrolysis

The fuels that can be used to provide chemical energy to assist with the electrochemical splitting of water include coal (raw or processed to remove sulfur and ash impurities), carbon from biomass sources, methane, methanol, ethanol and some other organic materials, such as dimethyl ether (DME) and ethylene glycol. In terms of cost, coal or carbon from biomass are the most effective fuel sources available in large quantities, and costs increase for hydrocarbon fuels.

Carbon fuels can be used in both low and high temperature electrolysis systems. Methanol and ethanol are more suitable for low temperature (< 80 °C) electrolysis systems, while methane is more suited for high temperature electrolysis cells. Methane is a major constituent of NG (typically > 92%) and is available in reticulated form in most developing countries. Alcohols and carbon are easily transportable fuels; thus the water electrolysis process to generate hydrogen can be carried out at distributed or demand centres. Obviously, coupling of the electrolysis process with renewable energy for low temperature electrolytic processes would offer environmental benefits, with electricity input provided by solar photovoltaic or wind, for instance. Similarly, for high temperature electrolytic processes in addition to renewable energy waste thermal energy from industrial processes or solar thermal plants can offer further environmental sustainability.

Coal, despite being the most obvious and low-cost option to assist with water electrolysis, is composed of complex aromatic clusters with inorganic constituents containing a large range of species which varies significantly for different coals. Fig. 1 is an example of molecular model

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