



Intensified hydrogen production and desulfurization at elevated temperature and pressure during coal electrolysis

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ABSTRACT

Elevation of temperature and pressure was used as an effective way to reduce the electrochemical resistance and improve the liquid mass transfer during the coal electrolysis without membrane, thus enhancing the desulfurization and hydrogen production. Results indicated that the high temperature and pressure have effectively reduced the electrochemical resistance (R_{ct}) and solution resistance (R_s) for coal electrolysis. The oxygen evolution reaction (OER) and hydrogen evolution reaction (HER) potentials for coal-water slurry electrolysis was obviously seen to increase with increasing temperature and pressure. What's more, the difference between the potential of OER and the equilibrium potential of OER (overpotential) also increased with the intensified condition, which was defined as the metastable zone width (MZW), indicating that HO^\bullet formation was improved by the high temperature and pressure. As expected, the elevation in temperature and pressure has successfully resulted in the decrease in cell voltage and the increase in coal desulfurization during the electrolysis.

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1. Introduction

Water electrolysis is a reliable and efficient method in the production of hydrogen [1], but there are many barriers preventing the development of this technology, such as the high power consumption resulted from the slow mass transfer in liquid phase and the high electrochemical resistance [2]. Therefore, it is necessary to reduce the power consumption during water electrolysis for hydrogen production.

To reduce the power consumption of water electrolysis, a number of methods have been proposed and developed, such as using different (noble) metal oxides as electrode materials [3–7], employing different types of electrolysis cells [8–10], and using new electrolytes [11]. It was noted that PEM water electrolysis was

also a kind of highly efficient approach for hydrogen production [12,13]. Besides, great efforts were also made to enhance the reaction rate, such as introducing ultrasound [14], super gravity field [15] and even square wave potential [16].

All these methods could more or less enhance the electrode reaction and mass transfer in the liquid phase. In other words, the splitting rate of water was improved, and the mass transfer of liquid phase was accelerated, thus achieving the reduction in the energy consumption for water electrolysis [17,18] and the increase in the current efficiency. However, some methods such as using ultrasonic wave [19] and super-gravity field [20] could only enhance the mass transfer rate of ion from electrolyte to electrode, but unfortunately failed to facilitate the electrode reactions.

It has been reported that the current efficiency of water electrolysis at 240 °C and 370 MPa, was up to 98.7% [2]. The enhancement in temperature and pressure could potentially improve the current efficiency during the water electrolysis [21]. With increasing temperature and pressure (especially, to achieve zero boundary state), the water dissociation constant (K_w) would increase obviously and produce more OH^- and H^+ . Under this circumstance, water could be highly miscible with nonpolar gases

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such as hydrogen and oxygen [22], and the reactants exhibited very high diffusion coefficients [23]. These advantages applied in the electrolysis cell would certainly increase the ionic strength of the electrolyte and reduce the mass transfer resistance of surface chemical reactions. For the electrolysis system under high temperature and high pressure, the chemical reaction energy barrier can be overcome with relative ease [24,25]. Therefore, the use of high temperature and pressure system can not only promote the HER of the cathode [26], but also has a great influence on the OER of the anode [27].

Table 1
Analysis of composition of high-sulfur coal.

Industrial analysis wt%			Elemental analysis wt%					
M _{ad}	A _{ad}	V _{ad}	FC _{ad}	C _{ad}	H _{ad}	O _{ad}	N _{ad}	S _{ad}
4.63	11.27	29.77	54.33	65.82	4.28	9.93	1.03	3.04

Coal electrolysis was efficient and energy-effective for hydrogen production [28], compared to water electrolysis [29]. More importantly, it could simultaneously achieve coal desulfurization and demineralization [30,31]. The coal desulfurization during electrolysis was actually the reaction between the sulfur and the active oxidants, which could indirectly reflect the ease at which the carbon was oxidized in the same reaction environment [32–34]. The coal electrolysis could change reactions (ΔG) on anode and hence reduce the internal resistance of the electrolysis cell [35]. OER was changed into sulfur element oxidation reaction, resulting in the decrease in the cell voltage for water electrolysis. Generally, the indirect oxidation process was predominant for coal electrolysis on anode, which could be expressed by following reactions in alkaline solution.

Direct electrolysis oxidation desulfurization reaction [14,36]:

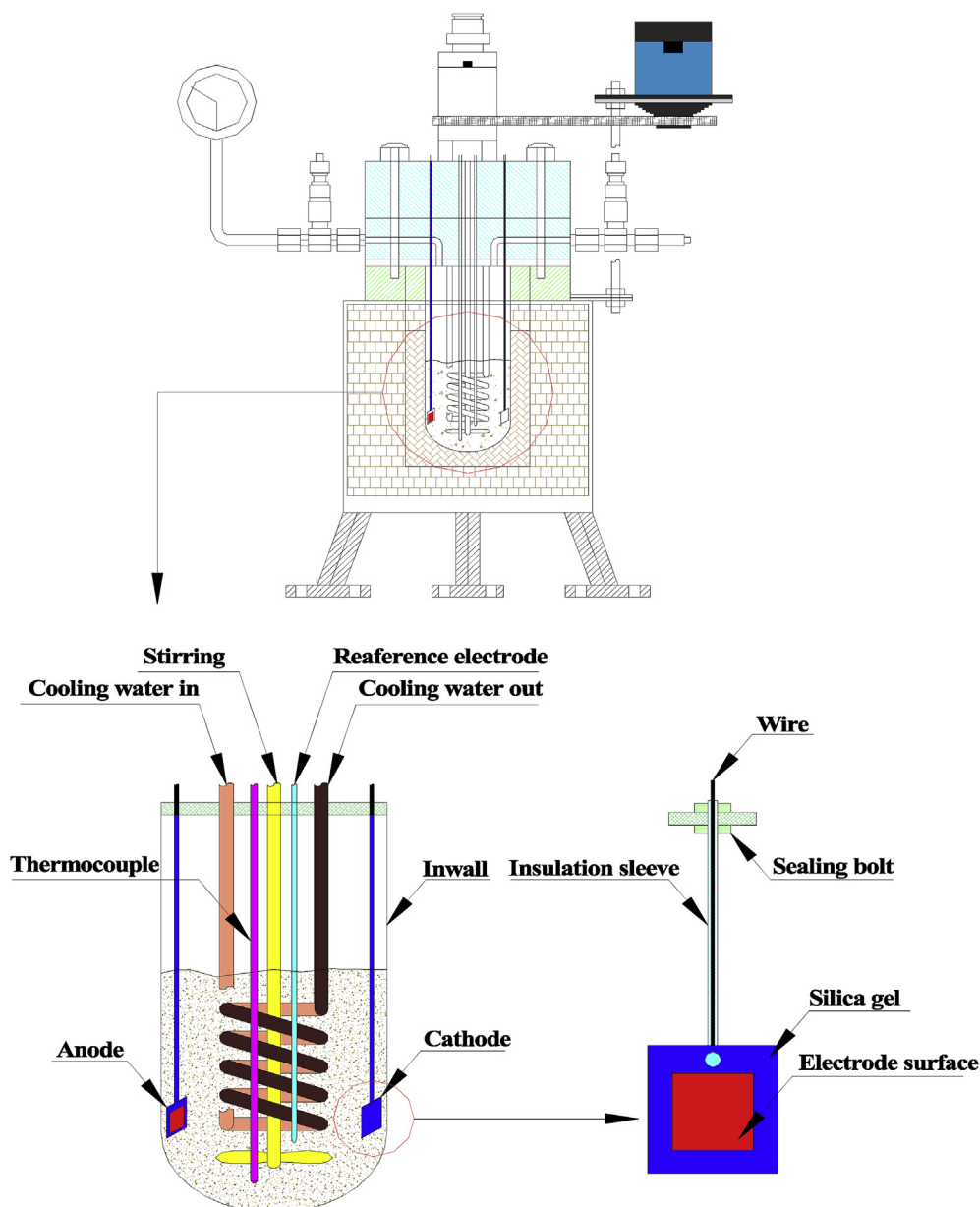
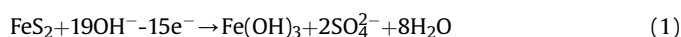


Fig. 1. Schematic diagram of high temperature and pressure electrolysis cell.

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