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Opportunities of integrated systems with CO_2 utilization technologies for green fuel & chemicals production in a carbon-constrained society

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ABSTRACT

With the increasing trend of global climate warming, effective control of CO_2 emission has drawn greater attention in the recent years. This has led to a significant challenge on the utilization of high carbon-containing resource such as coal. One of road-maps is the low-carbon and efficient utilization of coal-based resource with the integration of clean or renewable energy resource such as natural & unconventional gas, wind, solar, and even nuclear energy. To meet the demand of low-carbon solutions, the current focus is on the processing of carbon resource into green chemicals and fuels in terms of proposed concepts, one is to minimize the $C \rightarrow CO_2$ reaction during the processing steps, the other is to recycle CO_2 via chemical transformation or the relevant carbon cycle. Based on the concepts above, a series of low carbon conversion technologies are analyzed and discussed for four integrated systems, including (1) integration of natural gas with coal to produce fuels & chemicals, (2) coupling of nuclear energy with coal or CO_2 processing, (3) CO_2 methane reforming for fuel/chemical production, and (4) integration of solar energy with high temperature CO_2/H_2O splitting. The results indicate that the integration of fossil resource with non-carbon energy will reduce the CO_2 emission significantly; particularly when using carbon in the form of CO_2, zero carbon emission can be achieved.

1. Introduction

The process of carbon cycle in nature influence the climate and environment change on earth [1–5,7]. In the atmosphere carbon exists in various forms, in which carbon dioxide (CO_2) is the main contributor for the global climate change due to the greenhouse effect. The carbon cycle describes the movement of carbon as it is recycled and reused throughout the biosphere. The plants from land and ocean absorb CO_2 from the atmosphere, and then return to the atmosphere through biological or geological processes and human activities in the form of carbon dioxide. Annually, there are billions of tons of carbon rapidly cycling between the terrestrial atmosphere and ocean [6]. However, this balance has been broken by the rapid development of human society. CO_2 emission produced by the human activities has far exceeded the capacity of nature transformation. As stated on the IPCC fifth assessment report, recent anthropogenic emission of greenhouse gases are the highest in history, and the human influence is the main contributor to the climate change [1].

According to BP world energy statistics, in the year 2014, global CO_2 emission has reached about 9.68 billion tons carbon [8], 60% of which has emitted to the atmosphere. Most of the remaining CO_2 went to the oceans and only a few was taken up by the terrestrial biota. However, the nature uptake capacity of CO_2 is about 3 billion tons carbon per year, and in order to maintain the balance, CO_2 emission needs to reduce by 60–70%, ca. 5–7 billion tons carbon per year [5]. The IEA indicates the 70% of CO_2 emission come from fossil fuel combustion, in which the coal, oil and natural gas occupies 44%, 35% and 20% respectively [9,10]. According to the forecast from world energy council (WEC) [11], the fossil fuel will remain as the major form of energy resource and dominate the global energy supply until the year of 2050 (Fig. 1). Therefore, clean and efficient utilization of fossil fuels will be the pathway to attain sustainable and low-carbon future energy.

As for the carbon cycle in the process of utilizing fossil resource, it obviously covers the processing of fossil fuel feedstock to power, fuels and chemicals, as well as the capture and reuse of CO_2. Among

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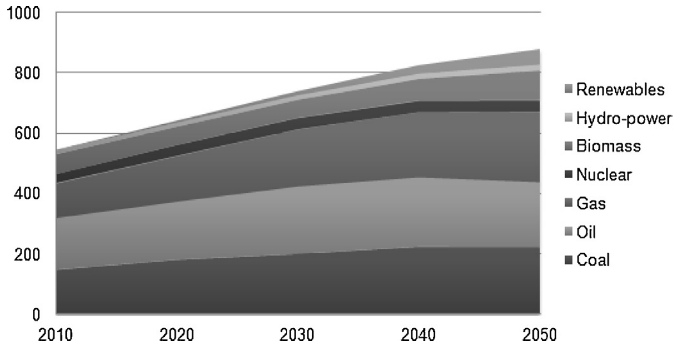

Fig. 1. Energy supply in 2050 as WEC's prediction (10^{18}J/a) [10].

them, carbon acts as the energy carrier and then terminates in the form of CO_2. In order to maintain the carbon equilibrium, any CO_2 produced from the other process rather than energy utilization has to be minimized, such as the CO_2 emission during the processes of energy conversion [12]. In the meantime, recycling of CO_2 also plays a very important role to maintain the carbon balance [13–15].

In the process of coal to methanol, for instance, the *H*/*C* ratio of the output from coal gasification is relatively low, and water gas shift reaction must be carried out to increase the *H*/*C* ratio. As a result, more than 50% of the carbon were converted to CO_2 [16–19]. From the view of carbon balance, such a process had relatively low carbon utilization efficiency, which is about 30% with the system energy efficiency of ca.45%. With an emphasis on green carbon energy, it is inevitable to consider about the improvement of carbon utilization efficiency at a reasonable energy efficiency for the carbon conversion process. Thus, the present discussion focus on the analysis of efficient utilization of fossil resources with low carbon emission, together with the relevant carbon cycle. In related to the chemical processing and utilization of fossil resource, it is important to minimize the reaction $C \rightarrow CO_2$ occur in the fuel or chemical production processes. On the other hand, capture and transformation of CO_2 into useful chemicals or fuels will be another major issue.

Coal gasification: $3C + O_2 + H_2O \rightarrow H_2 + 3CO$ (1)

Water gas shift: $CO + H_2O \rightarrow CO_2 + H_2$ (2)

Reverse water gas shift: $CO_2 + H_2 \rightarrow CO + H_2O$ (3)

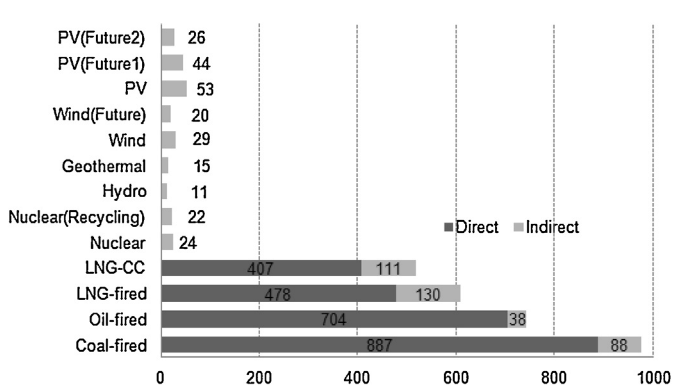

Fig. 2. Life-cycle carbon emission of power generation system (gCO_2/kWh) [20].

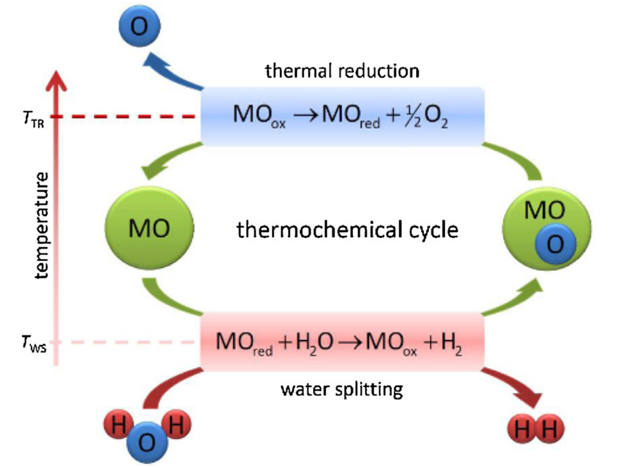

Fig. 3. General schematic of the two-step thermochemical cycle for water splitting. (MO denotes a metal-based redox material).

2. Development of low-carbon technology

The reactions involved in the energy processing usually require external energy inputs to achieve the required reaction conditions. For example, the processes of water electrolysis and high temperature splitting of CO_2/H_2O require intensive electricity or heat to sustain the endothermic reactions. Until now, 70% of global electricity supply comes from fossil resource (coal, oil and natural gas), 48% of which comes from coal combustion [10]. The amount of CO_2 emission associated with electricity generation from fossil resource is 10–20 times (see Fig. 2) of that from renewable energy and nuclear energy, according to the report by Hondo [20]. Similarly, most of the heat supply are comes from combustion of fossil fuel, which is another large contributor to CO_2 emission. The fundamental realization of CO_2 mitigation can only be achieved if energy supply for the fuel/chemical processing are replaced by renewable and clean energy rather than fossil based energy [17–19,21,22]. Hydrogen is an essential element in the energy conversion processing. The green production of hydrogen which assisted by nuclear or renewable energy is key to realize the fundamental reduction of carbon emission. Moreover, the recycling and transformation of CO_2 into useful fuels/chemicals is another important approach to reduce the CO_2 emission.

2.1. Typical low carbon technology with less $C \rightarrow CO_2$ reaction

Compared with fossil fuel to hydrogen production process, the use of nuclear and renewable energy are sustainable methods for hydrogen production by water electrolysis with high purity and green process [23–25]. There are three main types of water electrolysis based on the electrolyte used in the cell, including alkaline water electrolysis system, solid polymer electrolysis (SPE) system and solid oxide electrolysis cell (SOEC) system [26,27]. Due to the high electrolysis temperature of SOEC (800–950 °C), high temperature gas-cooled reactor (HTGR) is generally used for power and high-temperature heat supply [28–30]. It has been reported, total hydrogen production efficiency of HTGR-driven high temperature electrolysis using SOEC can be up to 55% which is 1.5 times of SPE electrolysis system and 2 times of alkaline water electrolysis system [31]. Therefore, the nuclear assisted high temperature water electrolysis is feasible to achieve zero carbon emission and practical for the large scale production of hydrogen.

The transformation of syngas to hydrocarbons, known as Fischer–Tropsch (F–T) synthesis, is another approach to sulphur- and nitrogen-free liquid fuels, and then more environmental