Chapter

Chemical Looping Combustion Power Generation System for a Power-to-Gas Scheme

Muhammad W. Ajiwibowo, Arif Darmawan and Muhammad Aziz

Abstract

Renewable energy provides a quick win solution for global warming, but it comes with drawbacks. Renewable sources such as solar and wind are not available for continuous use; thus, intermittency of electric power generation is an issue. Fluctuation of electricity production could damage the grid. Throughout the years, researchers have come up with solutions to solve this problem by storing the excess electricity via an energy storage system. One of the most efficient options is through solid oxide electrolysis cell (SOEC) to produce H2. In itself, H2 contains a lot of energy and can be converted to electricity via combustion or fuel cell. Therefore, storing electricity in the form of H2 could prove to be effective. Energy storage systems such as power-to-gas may provide a clean and efficient way to store the overproduced electricity. In this work, a power-to-gas system coupled with a chemical looping combustion combined-cycle system is proposed to provide base and intermediate load power from the unused electricity from the grid. Enhanced process integration was employed to achieve optimal heat and exergy recovery. This chapter focuses on the design of a system consisting of a power-to-gas conversion method and a H2-powered chemical looping combustion power generation system.

Keywords: chemical looping combustion, solid oxide electrolysis cell, system modeling, power to gas

1. Introduction

Combustion of fossil energy sources for industrial processes around the world contributes massively to the creation of greenhouse gases (GHG) and mainly CO2. This has been a major problem worldwide as it directly increases the pollution level and the likelihood of the earth’s increase in temperature. This led the international community to investigate ways to prevent this phenomenon [1]. Besides, fossil fuels are inevitably bound to be depleted in the future [2]. As of right now, as much as 84% of the world energy consumption is still fossil fuel driven, less than what the previous year had (85%); even so, it is still very high. Without a radical transformation, fossil fuel will still be the majority of energy source in the foreseeable future. This condition necessitates scientists and engineers to provide a sound solution. Efforts to reduce the fossil fuel usage and transition to sustainable energy
sources remain a challenge for scientists and engineers alike. Alternative renewable and clean energy technologies are actively being developed now [3, 4]. Fossil fuel combustion for power generation and industrial processes around the world is a key contributor to CO₂ emissions and had caused the earth’s temperature to increase ever since the industrial revolution. This brought the international community to implement emission regulations and policies to mitigate GHG effect on the earth while transitioning to sustainable alternatives to fossil fuels [1]. This is also supported by the fact that fossil fuel reserves are depleting due to massive use around the world [2]. Right now, as much as 84% of the world energy consumption is still derived from fossil fuels, which is less than what the previous year had (85%). Without a radical change, fossil fuel will still be the key energy source in the foreseeable future. Thus, efforts to reduce the fossil fuel usage and transition to sustainable energy sources remain a big challenge. Alternative renewable and clean energy technologies are actively being developed now [3, 4].

It is widely agreed and practiced that the conversion of fossil fuels into energy is mainly through combustion. It is an extremely efficient process and very mature in terms of technology. Its development dates back to the seventeenth century when the first steam engine was introduced. Regardless of its convenience now, combustion processes release lots of GHG and harmful gases (e.g., CO₂, SOₓ, NOₓ). Even so, combustion technologies will still thrive as a primary contributor to the world’s power generation. This necessitates scientists to develop a combustion method that is also environmentally friendly. Many research and development efforts have been put into finding new combustion methods. One of the major drawbacks of traditional method for combustion utilizes air, where, upon reaction, NOₓ and sometimes SOₓ will form and will potentially cause health indications if inhaled over time. One of the most promising solutions for a clean combustion process is to react the fuel directly with pure oxygen, or simply called oxy-combustion. This method will only generate CO₂ and H₂O emissions. Although being clean and efficient, the drawback of this process is that it requires prior separation of oxygen from the air in the atmosphere, which directly increases cost and efficiency penalty for the power plant.

Moreover, one similar solution, the emission could just be removed completely if renewable energy is used. Even so, the drawback of renewable energy usage is the intermittency of its production. Thus, the development of renewable energy systems throughout the years has generated a new interest in energy storage technologies. It is a dominant contributor in a renewable energy system. Despite being sustainable and clean, it has been reported that the use of renewable energies from solar and wind power sources has caused burdens to the electricity grid. Energy storage technologies include H₂, batteries, flywheels, compressed air, ultracapacitors, pumped hydro, and compressed gas [5]. Energy storage could mitigate power variations, enhance the system’s flexibility, and provide a scheme where surplus electricity from the grid could be stored and dispatched on demand [6].

Driven by the same problem, overproduction of electricity is also being demonstrated in Indonesia where renewable energy is not widely available yet. Heavily reliant on fossil fuels, the country consumes as much as 87.5% of all its power generation right now [7, 8]. Furthermore, according to the National Electricity Company in Indonesia (PLN), by default, Java-Bali sector of Indonesia’s power generation generates as much as 6000 MWe excess electricity production, which is wasted due to lack of energy demand. Likewise, this indicates a strong need for energy storage option. It is widely considered that the conversion of fossil fuels into energy is mainly through combustion. The development of combustion processes for power dates back to the seventeenth century when the steam engine
was first founded. With all its convenient characteristics, combustion process releases lots of GHG and harmful gases (e.g., CO₂, SOₓ, NOₓ). Seeing that combustion technologies will remain as a primary contributor to the world’s power generation, many research and developments have been put into finding new combustion methods. One of the promising ideas for clean combustion is to react the fuel directly with pure oxygen, or simply called oxy-combustion. This method will only generate CO₂ and H₂O emissions. Although being clean and efficient, this method requires prior separation of oxygen from the atmospheric air, which may increase cost and cause efficiency penalty for the power plant.

Moreover, the GHG emission could just be removed completely if renewable energy is used. The development of renewable energy systems throughout the years have generated a new interest in energy storage technologies. It is a dominant contributor in a renewable energy system. Despite being renewable and clean, it has been reported that the use of renewable energies from solar and wind power sources have caused burdens to the electricity grid. Energy storage technologies include H₂, batteries, flywheels, compressed air, ultracapacitors, pumped hydro, and compressed gas [5]. Energy storage could mitigate power fluctuations, enhances the system’s flexibility and provides a scheme where surplus electricity from the grid could be stored and dispatched on demand [6].

By the same token, overproduction of electricity is also being demonstrated in Indonesia, where renewable energy is not widely available yet. Heavily reliant on fossil fuels, the country consumes as much as 87.5% of all its power generation right now [7, 8]. Furthermore, according to the stated-owned electricity company in Indonesia, PLN, by default, Java-Bali sector of Indonesia’s power generation generates as much as 6000 MWe excess of electricity production, which are wasted due to lack of energy demand. Likewise, this indicates a strong need for energy storage option.

1.1 Hydrogen production and utilization

Hydrogen as an alternative energy source is predicted to have a powerful role in the low-carbon future [9]. It is long recognized as a sustainable fuel due to its favorable characteristics [10]. Besides that, hydrogen is greatly abundant on earth, albeit in its oxidized state (H₂O) [11]. Presently, H₂ is commercially produced using reforming technologies on hydrocarbon fuels. Researches for the production of H₂ have been a great interest for a long time. The routes to produce H₂ vary from chemically to electrochemically. Chemically, supercritical water gasification (SCWG) of biomass and syngas chemical looping (SCL) are among the most efficient ways to produce hydrogen [11, 12]. Compared to other types of gasification, SCWG utilizes more steam, thus promoting more reforming of the gases to produce H₂.

Moreover, from the electrochemical routes, many types of water electrolysis are considered to be effective for H₂ production by splitting water. Proton-exchange membrane (PEM) electrolysis and alkaline water electrolysis are the most used technology due to their maturity [13]. Typically, PEM electrolysis and alkaline water electrolysis have energy efficiencies of around 50–80%. Furthermore, solid oxide electrolysis cell (SOEC) is another type of electrolysis method that utilizes a high temperature of around 750–1000°C and 10–15 bar of pressure [14]. In higher temperatures, higher efficiency could be achieved due to smaller energy requirements.

The utilization of H₂ also varies from just energy storage to electric power production. Various H₂ energy conversion technologies are already generally understood, and many are already under commercial real-world developments
Fuel cells are among the best and are among the most efficient ways for energy conversion from H2 for electric power production. Integration of fuel cells with other technologies remains a challenging puzzle. Various integrations of fuel cell and other systems are currently being investigated. Generally, fuel cell is very versatile in terms of fuel types and various operating conditions. Nevertheless, one of the major drawbacks of fuel cell is that the membrane used in the cells may have a relatively short lifetime and, hence, could potentially add costs to fuel cell systems.

Numerous efforts have been made to integrate fuel cells, especially SOEC with other energy conversion technologies. Cinti et al. proposed and investigated an integrated SOEC and Fischer-Tropsch system to produce methane from surplus renewable energy. Energy and exergy evaluation of an SOEC-methanation system is also evaluated by Luo et al. Kezibri et al. also modeled a power-to-gas system for oxy-combustion power generation. But they utilized and considered a proton-exchange membrane-based electrolyzer, which is heavily reliant on the membrane and did not further evaluate the possibilities of system and heat integration. All these efforts can prove high potential of hydrogen energy utilization in the future. Unfortunately, there is still no significant effort made to provide an efficient energy system utilizing SOEC technology for hydrogen and, subsequently, power.

1.2 Chemical looping combustion power system

Chemical looping combustion (CLC) is a new and leading-edge energy conversion method that utilizes metal oxides or otherwise called oxygen carriers to oxidize the fuel instead of atmospheric air. It uses two reactors, namely, the reducer and the oxidizer. Mixtures of metal oxide and some inert solids for heat dilution are being circulated throughout these two reactors. The process starts when fuel is oxidized in the reducer reactor; afterwards, the reduced metal oxide will be re-oxidized by air in the oxidizer while also generating heat in the process. These processes are being repeated in a looping fashion. There have been many reports on the use of various types of fuels, including hydrocarbon and biomass. Typically, exhaust gas containing CO2 and H2O will be generated from the fuel oxidation, and CO2 will be easily separated by condensation. Inherent CO2 separation and storage are possible in this system due to the separate combustion processes. Furthermore, if H2 is used, the combustion product will be just H2O. Thus, this combustion technology has a high potential as a zero emission power generation system due to its favorable characteristics, namely, the inherent separation capability for CO2. As a comparison, this system could achieve the same advantage that an oxy-combustion power plant has without the need to separate air first with an air separator unit (ASU).

Thus, this paper tries to focus on the effort to propose an efficient energy system which comprises of SOEC and chemical looping combustion (CLC) combined cycle power generation to produce baseload power from hydrogen as a sustainable zero emission energy source. The produced H2 is fed to the CLC power generation system where it is mixed with natural gas. This way, environmental burden caused by GHG can be decreased as we are producing less CO2 compared to traditional natural gas-fired power generation system. Detailed process integration and the calculation are discussed in Sections 2 and 3. Besides that, parameters and operation conditions are evaluated to further improve the system's energy conversion efficiency and its key design parameters.

This paper focuses on the effort to propose an efficient energy system that comprises of SOEC and chemical looping combustion (CLC) combined cycle power
generation to produce base and intermediate loads power from both natural gas and renewable energy-based hydrogen. The produced \( H_2 \) from renewable energy source is fed to the CLC power generation system where it is mixed with natural gas. By this way, environmental burden caused by GHG can be decreased as the process is producing less \( CO_2 \) compared to traditional natural gas fired power generation system. In addition, the inherent fluctuation characteristic of renewable energy can be stabilized, producing a stable and reliable power generation to the grid. Detailed process integration and the calculation is discussed in Sections 2 and 3. Besides that, parameters and operation conditions are evaluated to further improve the system’s energy conversion efficiency and its key design parameters.

2. Proposed integrated system

2.1 Integrated SOEC and chemical looping combustion power generation system

Figure 1 depicts a simplified process flow diagram for the proposed system. The system itself consists of an SOEC as the \( H_2 \) producer module and a chemical looping combustion combined cycle (CLCCC) as an electric power generation module. Theoretically, the CLCCC could act as the main electric power generator providing a stable electrical power output that is suitable for base and intermediate loads. The \( CO_2 \) emitted from the combustion process is basically separated. This leads to low \( CO_2 \) emission and potentially zero \( CO_2 \) emission if \( H_2 \) is used. For this system, a dual fuel system is considered where natural gas is considered, while the \( H_2 \) produced from the electricity from renewable energy sources or surplus electricity from the grid becomes additional fuel. Theoretically, the produced \( H_2 \) is consumed without being stored; therefore, the flow of natural gas to CLCCC decreases accordingly. Although, a storage infrastructure could also be considered for \( H_2 \) in such system, it is not considered for this study. Hence, the generated electric power from the system is assumed stable, without being influenced by the fluctuation that comes with renewable energy sources and grid surplus electricity.

Overall, surplus electricity from the grid is used to convert \( H_2O \) into \( H_2 \) and \( O_2 \) via the SOEC process. Afterwards, as described in the previous part, the \( H_2 \) is directed and fed directly to the CLCCC module. On the other hand, additional \( O_2 \) is also being fed into the CLCCC power system along with air, which potentially leads to higher combustion temperature. Table 1 describes the main assumptions and parameters used for the developed CLCCC system.

In order to achieve highest energy efficiency for the system, enhanced process integration methodology is utilized. This approach primarily focuses on heat and exergy recovery in the system via heat exchanger integration and compression [20, 21].

![Figure 1](http://dx.doi.org/10.5772/intechopen.85584)

*General overview of the integrated system.*
Waste heat from hot downstream processes is utilized to support the heat requirements of upstream processes recuperatively. Exergy is also elevated in cold streams via compression. It is a proven methodology that has been demonstrated by various works for producing electricity or hydrogen from various sustainable sources, especially biomass [9, 11, 22].

Basically, the CLCCC is somewhat similar to a traditional combined cycle with a gas turbine, but the combustor in such system is replaced by two chemical looping combustion reactors that act as the heat source for the downstream turbines. As opposed to the traditional method, the produced fuel gas that is rich in CO₂ could be directly separated as it does not produce other by-products except CO₂ and H₂O. Thus, the separation of CO₂ is significantly less energy intensive than the traditional separation method. The schematic diagram of CLCCC is shown in Figure 2. In a dual fuel scenario, H₂ and CH₄ (natural gas) are considered to be fuels for this system, with the key assumption of a reactor design that would support the use of these two fuels. Two fuel gas streams coming out of the CLC process, namely, the reducer gas and the oxidizer gas, are expanded via the reducer turbine (RT) and the oxidizer turbine (OT). Afterwards, the CO₂-rich stream leaving the RT is directly separated by condensation and then compressed and stored.

Table 1. Details on the parameters and assumptions used in the CLCCC.

| Component/system                       | Unit | Value         |
|----------------------------------------|------|---------------|
| Solid composition                     |      | 70% metal oxide |
|                                        |      | 15% SiC       |
|                                        |      | 15% Al₂O₃     |
| Generator efficiency                  | %    | 98            |
| Compressor isentropic efficiency      | %    | 90            |
| Turbine isentropic efficiency         | %    | 90            |
| Fuel flow rate                        | kg/s | 6             |
| OT inlet temperature                  | °C   | 1400          |
| RT inlet temperature                  | °C   | 800–900       |
| ST inlet temperature                  | °C   | 700           |
| Operating pressure                    | Bar  | 15–35         |
| ST inlet pressure                     | Bar  | 250           |

Figure 2. CLCCC power generation system.
the other hand, the high-temperature gas leaving the OT is used to generate steam for generating more power via a steam cycle.

### 2.2 Solid oxide electrolysis cell for H₂ production

As discussed in the previous section, SOEC is regarded as one of the most efficient electrolysis processes to produce pure H₂ [23]. For the purpose of this study, SOEC is considered favorable if it is used for power-to-gas energy storage. In this case, the input electricity used for the SOEC is the surplus electricity from the grid (especially due to surplus from renewable energies) or directly from renewable energy sources. Afterwards, the generated H₂ will be stored temporarily or can be directly integrated with the CLCCC system described before. The SOEC parameters in this study are based on the research conducted by Udagawa et al. [23]. The detailed parameters are provided in Table 2.

| Component/system                  | Unit     | Value |
|-----------------------------------|----------|-------|
| Power input                       | MW       | 350   |
| Produced hydrogen                 | Nm³/h    | 87,803|
| Produced oxygen                   | Nm³/h    | 43,901|
| Operating temperature             | °C       | 750   |
| Operating pressure                | Bar      | 10    |
| Efficiency                        | %        | 88    |

Table 2. Assumptions for the SOEC used in the model.

3. **Process modeling and calculation**

For the purpose of system, mass, and energy balance simulation, ASPEN Plus V8.8 from Aspen Technology, Inc., is utilized in this study. Key assumptions made for this model are listed in Table 1 that are primarily taken from other experimental and numerical researches. The operating conditions are chosen based on other literatures [23–25]. Key thermodynamic assumptions are as follows: (i) ambient temperature is set to 27 °C; (ii) no heat loss is assumed; and (iii) air is assumed to contain 79% mol N₂ and 21% mol O₂. RStoic reactor blocks are used to model the reducer and oxidizer reactors in ASPEN Plus. Simplistically, in the reducer, the metal oxides will be reduced by the fuels, which are H₂ and CH₄, and then circulated to the oxidizer where it is re-oxidized by air. The operating reactors are assumed to be an entrained flow type for the oxidizer, and a moving bed type is used for the reducer.

To identify different parameters used in this study, three types of metal oxides are evaluated for the CLCCC process. Two of the most studied oxygen carriers, nickel oxide (NiO) and iron oxide (Fe₂O₃), are each utilized for the CLCCC process. Additionally, CaSO₄, also known as gypsum, is also utilized and evaluated in this study due to recent interests for this material as an oxygen carrier due to its favorable chemical characteristics [26]. All of these metal oxides have many distinctive characteristics, such as resistance to elevated temperatures, high oxygen concentration, and so on that could provide extra benefit and efficiency boost to the CLCCC system. Table 2 provides the description of the metal oxides used in this study.
And then, for parametric purpose, different H$_2$ and CH$_2$ mix flow rates are considered. It is aimed to simulate different intermittent or fluctuating renewable energy source used to generate H$_2$ in the SOEC. The key assumption is that the heat rate for the CLCCC is assumed to be the same for each different mixture. The base case is 6 kg/s of H$_2$. And then, for the metal oxide mixture, inert solids are also considered as heat diluents and heat carriers. The mass fraction is assumed to be 70% metal oxide and 30% inert materials, which consist of 15% SiC and 15% Al$_2$O$_3$ as suggested by Fan [25]. Figure 3 shows the detailed process flow diagram of the proposed system (Table 3).

As described in previous parts, the process begins with the fuel produced in the SOEC entering the CLCCC along with natural gas. The mixture is then directly combusted in the reducer. The fuel gases leaving the reducer and oxidizer are expanded in the RT and OT, respectively, for electricity production. The CO$_2$ leaving RT is separated and compressed up to 200 bars for storage. Besides that, the fuel gas leaving the OT is utilized for heat addition of air and water streams. On the other hand, the heat in reducer gas is also used to heat the water feed stream in HX-1 and compressed air in HX-2. And then, HX-2 is also utilized to cool down the compressed air coming from C2 to reduce additional compression work in downstream processes.

The system’s total energy efficiency is defined and calculated as follows:

\[ W_{Gen} = W_{RT} + W_{OT} + W_{ST} \]  

\[ W_{Used} = W_{Compressors} + W_{Pump} + W_{Auxiliary} \]  

\[ \eta_{tot} = \frac{W_{gen} - W_{used}}{LHV.m_{fuel}} \]  

where, $W_{Gen}$, $W_{RT}$, $W_{OT}$, and $W_{ST}$ are the total work obtained from the system, works obtained from RT, OT, and ST, respectively. In addition, $W_{Used}$,
$W_{\text{Compression}}$, $W_{\text{Pump}}$, and $W_{\text{Auxiliary}}$ are the total consumed work by the system, works consumed by compressors, pumps, and auxiliaries, respectively. Finally, the total energy efficiency $\eta_{\text{tot}}$ is defined as the ratio of net produce power to the total calorific value of the fuels, including $\text{H}_2$ and $\text{CH}_4$.

### 4. Results and discussion

The parametric analysis has been done to provide a better understanding of the key design parameters of the SOEC-CLCCC system. First of all, as mentioned in the previous section, $\text{CaSO}_4$, $\text{Fe}_2\text{O}_3$, and $\text{NiO}$ are each adopted and evaluated in the CLCCC system as oxygen carrier. Afterwards, different operating pressures from 15 to 35 bars, with an interval of 5 bars, are simulated for each system. Furthermore, different percentages for the $\text{H}_2$ used as fuel are also being investigated.

For the case of $\text{CaSO}_4$ as the oxygen carrier, the results are presented in Figure 4. The effects of different pressures and fuel mixtures are evaluated in the graph. The main characteristics of this particular oxygen carrier are high oxygen transport capacity, high temperature resistance, and highly exothermic reactions with the fuels used. Generally, the main driver for efficiency is the different fuel mixture. The highest energy efficiency obtained is 53%, with full $\text{H}_2$ feed. The efficiency decreases along with decreasing $\text{H}_2$ use. Basically, more $\text{CH}_4$ is required with each incremental reduction of $\text{H}_2$. Thus, more compression duty is required for the fuels due to higher flow rate. By the same token, lower pressures are more favorable for this case due to high compression work required. Thus, 15 bars achieve the highest efficiency of 54%.

Afterwards, when $\text{Fe}_2\text{O}_3$ is used as oxygen carrier, the highest energy efficiency obtained by the system rises to about 56%. The relationship is depicted in Figure 5. Basically, the trend is similar to the $\text{CaSO}_4$ system. When the $\text{H}_2$ percentage decreases, the energy efficiency decreases accordingly to about 45–47%. And then, lower pressures tend to be more preferable to achieve higher energy efficiency. This
oxygen carrier has the lowest oxygen carry capacity compared to the other two oxygen carriers, but it has the highest melting temperature.

Furthermore, Figure 6 shows the relationship of the operating pressures and the hydrogen percentages to the energy efficiency when NiO is used as oxygen carrier. In this case, the achievable maximum energy efficiency is about 56%. When H₂ percentage is reduced, the energy efficiency of the system further decreases to about 45–47%. Moreover, when larger amount of CH₄ is supplied, larger energy is required for CO₂ compression. NiO as an oxygen carrier has the second highest oxygen carry capacity and melting temperature compared to the other two oxygen carriers used in this study. These differences in efficiencies are driven by the difference of the heat released by the reactions that exist in the reducer and oxidizer.

Generally, from process modeling, operating pressure of the CLC system plays a significant role on changing the system’s power intake and production. Process modeling suggests that the highest compressor work is consumed for the air compression process. Due to lower heating value of CH₄ compared to H₂, the system
energy efficiency decreases accordingly when larger amount of CH₄ is used as fuel. Besides that, the thermodynamic characteristics also played a significant role to determine the heat produced and requirement in the combustion system. Subsequently, in case of CaSO₄ is used as oxygen carrier, as the reaction of CH₄ with CaSO₄ is endothermic, the input energy is required. This is opposite to the H₂ that provides an exothermic reaction releasing a considerable amount of heat. From process modeling, it can be inferred that the highest amount of heat exchange occurs in the oxidizer, where the heat from the oxidizer is utilized to heat up the steam to a higher temperature.

Compared to other power generation systems, the proposed system does not require additional process for air separation unit (ASU) and CO₂ separation process. Yet, the proposed system can achieve relatively high energy efficiency, which is similar to the energy efficiency of oxy-combustion power system that requires further CO₂ separation process and ASU.

**Figure 6.**
*System energy efficiency vs. operating pressure for the NiO system.*

**Figure 7.**
*Total solid flow rate and the CO₂ production for each system.*
the CLCCC system, because air that is usually used for heat dilution could be replaced totally with solids. For further optimization of the system, excess air or excess solids can be considered as temperature reduction agents in the oxidizer. Higher amounts of solids circulated will require bigger reactors and more solid control infrastructure, and higher amount of air flow would require more compressor work.

5. Conclusion

A clean and efficient energy system to utilize efficiently hydrogen produced from renewable energy or surplus electricity is proposed. The proposed system is based on the chemical looping combustion using CH₄ as the base fuel. The system consists of SOEC for hydrogen production, chemical looping combustion, and combined cycle for power generation. The cleanliness of the developed system is promising as it can separate CO₂ directly via condensation due to the clean combustion process of CLC. A high energy efficiency of about 56% can be obtained. The results of this study are useful for further improvement and developments toward an actual CLC power generation system.

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Chemical Looping Combustion Power Generation System for a Power-to-Gas Scheme
DOI: http://dx.doi.org/10.5772/intechopen.85584

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