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Low-carbon power generation cycles: the feasibility of CO_2 capture and opportunities for integration

Wojciech M. Budzianowski*

Wrocław University of Technology,
Wybrzeże Wyspiańskiego 27, 50-370 Wrocław, Poland

Abstract

Low-carbon power generation is receiving increasing interest due to climate warming concerns. The present article analyzes three low-carbon power cycles. The focus is on the feasibility of CO_2 capture and opportunities for energy and mass integration. The first power cycle is a zero-carbon solid biomass fuelled multi-step gasification gas turbine power cycle involving multi-step solid biomass conversion, which is a more reversible process than one-step biomass combustion. The second zero-carbon coal-fired oxy-gasification steam chemical looping combustion gas turbine cycle benefits from: (i) improved cycle efficiency due to the increased reversibility of the chemical looping combustion process, (ii) cycle mass and energy integration due to the several recirculation loops involved, and (iii) extremely high CO_2 capture rate due to the purity of the CO_2/H_2O mixture achieved at the outlet of a syngas reactor. The last power cycle - a biogas fuelled oxy-reforming fuel cell cycle - is superior in terms of the feasibility of CO_2 capture, i.e. CO_2 is captured from CO_2-enriched streams, and due to the utilization of renewable biogas, negative net CO_2 atmospheric emissions are achieved. It is concluded that high CO_2 capture rates are feasible from pressurized CO_2-enriched streams comprising either water or hydrogen, thus necessitating oxy-fuel power cycles. Opportunities for mass and energy integration are found to be greater in systems involving closed mass and energy recirculation loops. The discussions also emphasize that low-carbon power cycles could achieve minimized exergy losses by applying more reversible energy conversion processes.

Keywords: Low-Carbon Economy, power generation, CO_2 capture, integration, reversibility.

1. Introduction

A Low-Carbon Economy (LCE) is an economy which has a minimal output of greenhouse gases into the biosphere. The means for achieving LCE are: renewable energy, nuclear energy, energy efficiency and carbon capture and sequestration (CCS). The end product of most power generation technologies involving carbonaceous fuels is CO_2, which is released to the atmosphere. This power generation-derived anthropogenic CO_2 accumulates in the atmosphere in a process which is practically irreversible in the modern era since photosynthesis-based natural processes are very slow and due to industrialization and deforestation plants' potential for atmospheric CO_2 binding is now substantially lower than in previous centuries. Consequently, anthropogenic CO_2 emissions affect the equilibria of natural carbon cy-

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*Corresponding author
Email address: `wojciech.budzianowski@pwr.wroc.pl` (Wojciech M. Budzianowski)

cles and CO_2 concentration in the atmosphere is increasing at a rate of around 2 ppm year-1 currently reaching around 390 ppm. At this rate the Copenhagen Accord target of 450 ppm CO_2/+2 °C will be reached before 2040. This clear need to cut greenhouse gas emissions to the atmosphere is driving research and investment in low-carbon power generation technologies. The CO_2 generation intensity of renewable and nuclear electricities ranges from 10 to 50 kg CO_2 MWh^{-1}. In contrast, fossil fuel-fired power generators are characterized by substantially larger CO_2 emission of around 400 kg CO_2 MWh^{-1} for natural gas, 850 kg CO_2 MWh^{-1} for oil and 900 kg CO_2 MWh^{-1} for coal. Thus, the decarbonization of fossil fuel-based power generation necessitates CO_2 capture and sequestration (CCS). CO_2 capture can be realized by using a variety of processes such as reactive absorption or membranes. CO_2 sequestration can be achieved through, for example, geological storage or through some much more interesting options such as CO_2 utilization in chemical industries, fuel synthesis, algae cultivation, enhanced gas and oil recovery or enhanced methane coal-bed recovery. Sustainable and renewable energy sources such as hydro, wind, solar and geothermal can offer low-carbon power generation, but they have limited energy capacities and produce relatively costly electricity. Fossil fuels, on the other hand, are available in large quantities and are used to produce relatively cheap electricity. The main problem, however, are the atmospheric CO_2 emissions caused by the combustion of carbonaceous fossil fuels. It is thus desirable to use the primary energy of fossil fuels, such as coal or hydrocarbons, but with limited CO_2 emissions. Alternatively, power can be produced from renewable biomass-derived fuels. However, in this case additional technical measures are needed to decrease the costs of electricity from biomass-based power cycles. Those two important topics are thus addressed in this paper. The main idea behind pre-combustion CO_2 capture techniques [1] in relation to carbonaceous fuels is to oxidize carbon to CO_2 while transferring all of the fuel's chemical energy into H_2, which can be further oxidized without any atmospheric release of CO_2. This article analyzes the feasibility of CO_2 capture and opportunities for mass and energy integration of low-carbon power generation cycles. In the first power cycle CO_2 is not separated due to the use of biomass fuel and the focus is on energy integration. The second and the third cycles involve pre-combustion CO_2 capture and discussions relate to the feasibility of cycle decarbonization and to opportunities for mass and energy integration.

2. Low-carbon power cycles

The investigated power cycles utilize various fuels such as solid biomass, coal and biogas and a variety of power technologies such as gas turbines, steam turbines and fuel cells. Moreover, novel processes suitable for low-carbon power generation are included, i.e. torrefaction, oxy-gasification, chemical looping combustion and oxy-reforming.

2.1. Solid biomass fuelled multi-step gasification gas turbine (MSG-GT)

Biomass is an abundant, untapped and environmentally friendly renewable energy carrier. Sustainable solid biomass resources available for energy use include: (i) agricultural residues (straw, grasses), (ii) forest residues (from e.g. sawmills), (iii) biomass production on surplus degraded land, (iv) organic wastes and (v) energy crops. Currently, most large-scale biomass-based power technologies are direct combustion (via e.g. CFB boilers) and co-combustion with coal. However, direct biomass combustion suffers from low energy conversion efficiencies (around 20 %) due to high moisture content and thus low flame temperatures achieved. This problem is overcome in co-combustion but this means biomass must be transported to centralized coal-fired power plants over long distances, which substantially increases overall costs. Therefore, novel, more efficient biomass-based power cycles are needed.

2.1.1. Promising technologies for solid biomass fuelled power plants

Solid biomass can be converted to gaseous fuels by heating in a gasification agent such as oxygen/steam, air or CO_2. In a gasifier solid biomass is pyrolyzed to volatile tars and to char. The char is then simultaneously oxidized and reformed to produce syngas. Biomass gasification is well suited for biomass IGCC (BIGCC) with combustion of syngas in a gas turbine (as a topping cycle) while hot gases from the gas turbine are used for steam generation in a heat recovery steam generator (HRSG) to run a steam turbine (a bottoming cycle). Torrefaction is a mild pyrolysis process that improves the fuel properties of solid biomass. Biomass torrefaction is realized at temperatures below 300 °C in the absence of oxygen. The main products formed are torrefied biomass and volatilities - torrefaction gas. The torrefied solid biomass is characterized by low moisture content, high energy density and small volume. In addition to improvements in the ease of grinding/pelletization, long-term storage potential and reduced long-distance transportation costs are achieved [2]. The torrefied biomass is suitable for direct combustion, co-combustion and gasification. Pre-treatment of biomass via torrefaction prior to gasification enables the use of a pressurized entrained flow gasifier and thus higher gasification temperatures and higher gasification thermodynamic efficiency can be achieved.

2.1.2. Thermodynamic considerations for efficient design of solid biomass fuelled power plants

Practical design principles for efficient energy conversion in solid biomass fuelled power plants can be derived directly from the First and Second Law of Thermodynamics. Namely, the First Law of Thermodynamics (the conservation of energy) necessitates that a power system under design should minimize energy losses to the environment. This can be achieved by thermal integration measures such as (i) minimizing radiation losses (via e.g. heat recovery or insulation techniques) and (ii) minimizing losses in output streams (via e.g. heat recirculation [3], flue gas recirculation [4] or fuel pre-drying. More interesting power plant design insights arise from the Second Law of Thermodynamics (the quality of energy). Namely, in order to minimize exergy losses and thus entropy generation rates a solid biomass power plant should involve biomass conversion operations characterized by decreased irreversibility. In combustion, most exergy destruction occurs during thermal energy exchange between hot reaction product molecules and cold reactant molecules [5]. The fuel oxidation itself is relatively efficient. This effect arises from the fact that in combustion processes fresh cold substrates are in direct contact with hot products, which is very inefficient, since the maximal temperature is reduced and thus less work can be delivered. Further, if an energy conversion process can take place with sufficient degrees of freedom, the optimal process follows a trajectory of optimal thermodynamic states, a so-called highway in state space, which is characterized by constant local entropy production [6]. This power plant design principle known as the equipartition of entropy production is satisfied in many biological systems such as human lungs (see [6] in relation to fuel cell design), and thus it can inspire highly efficient design of power plants.

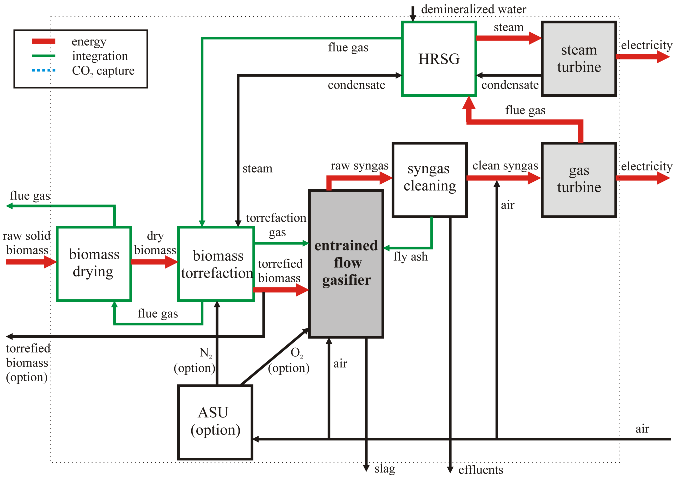

Figure 1: The flowsheet of the solid biomass fuelled power plant utilizing multi-step gasification gas turbine (MSG-GT) power cycle

2.1.3. Multi-step gasification gas turbine (MSG-GT) power cycle

In light of the aforementioned first- and second-law thermodynamic considerations one-step solid biomass combustion seems to be less efficient solution than multi-step solid biomass gasification. Namely, the multi-step solid biomass gasification technology can assure more reversible gradual fuel oxidation in multiple sub-units thus limiting thermal energy exchange between hot reaction products and cold reactants as well as enabling advanced energy integration.

Figure 2 presents a flowsheet of a power plant utilizing multi-step solid biomass gasification technology. Accordingly, the proposed multiple biomass conversion steps involve biomass pre-treatment processes such as drying and torrefaction followed by gasification and final syngas combustion in a combined cycle gas turbine. Gasification itself can be realized in a high-temperature air-blown entrained flow gasifier involving chemical quench via the injection of torrefaction gases, thus offering improved char and tar conversion even at low oxygen consumption as well as improved ash to slag fusion. Moreover, the proposed flowsheet is in accordance with the principle of equipartition of entropy production, i.e. entropy is produced more uniformly in all sub-units of the proposed power plant. The biomass conversion route according to the flowsheet from Figure 2 can be more energy efficient, cleaner and optionally it can enable to manufacture a new valuable renewable product - the torrefied biomass.

2.2. Coal fuelled oxy-gasification steam chemical looping combustion gas turbine (OG-SCLC-GT)

Chemical looping combustion can potentially offer increased energy conversion efficiency. The gain in efficiency is possible due to the enhanced reversibility of the two redox reactions. In conventional direct combustion, the release of a fuel's chemical energy occurs in a highly irreversible manner - departing considerably from equilibrium [5]. In contrast, in CLC, both redox reactions can occur almost reversibly and at relatively low temperatures. This can allow a power plant using CLC to approach the ideal work output for an internal combustion engine without exposing its components to excessive operating temperatures. Moreover, CLC facilitates CO_2 capture because the two redox reactions generate two intrinsically separated flue gas streams with one consisting entirely of CO_2 and H_2O. This gives CLC clear benefits when compared with competing carbon capture technologies, as the latter generally involve a significant energy penalty associated with either post combustion scrubbing systems [7] or the work input required for air separation plants. Therefore, CLC-based technologies can achieve high efficiency power generation with low energy penalty carbon capture. A steam chemical looping combustion (SCLC) method consists of oxidation and reduction processes, which are undertaken in two separate reactors. In the reduction step, the syngas is reacted with an oxygen carrier such as iron oxide to form carbon dioxide and water. After condensing the water vapor, the captured carbon dioxide stream can be sent to the sequestration sites. The reduced form of iron is re-oxidized in an oxidation reactor to its original form using steam and recycled back to the reduction reactor. In the fuel reactor the syngas is oxidized with iron oxide (magnetite) according to the following reactions:

$$Fe_3O_4 + 4CO \rightarrow 3Fe + 4CO_2 \tag{1}$$

$$Fe_3O_4 + 4H_2 \rightarrow 3Fe + 4H_2O \tag{2}$$

In the steam reactor the reduced form of the oxygen carrier (iron) is oxidized back by using steam to regenerate the iron oxide and to produce hydrogen according to the reaction:

$$Fe + 4H_2O \rightarrow Fe_3O_4 + 4H_2 \tag{3}$$

Hydrogen is then sent to a combined cycle gas turbine. Figure 2 presents the flowsheet of an oxy-gasification steam chemical looping combustion gas turbine (OG-SCLC-GT) power system.

A main advantage of the OG-SCLC-GT is that it can offer a CO_2 capture rate exceeding 99 % while maintaining efficiency comparable to other power generating/CCS technologies. The consumption of oxygen can be minimized in optimized gasifier designs [8]. The water recirculation loop (i.e. gasifier/CLC/condenser) minimizes water consumption, thus mass and energy integration is ensured. Coal pre-drying enables energy to be extracted from flue gases.

2.3. Biogas fuelled oxy-reforming fuel cell (OR-FC)

Biomass includes carbon entirely assimilated from atmospheric CO_2 during its growth via photosynthesis with the contribution of solar energy, since plants are unable to assimilate carbon from any other source. Hence, bioenergy processes that involve the utilization of biomass-derived biogas with simultaneous CO_2 capture are characterized by 'negative net CO_2 emissions' to the atmosphere [9]. Decarbonization of biogas is a much more attractive option than CO_2 separation from fossil fuel power

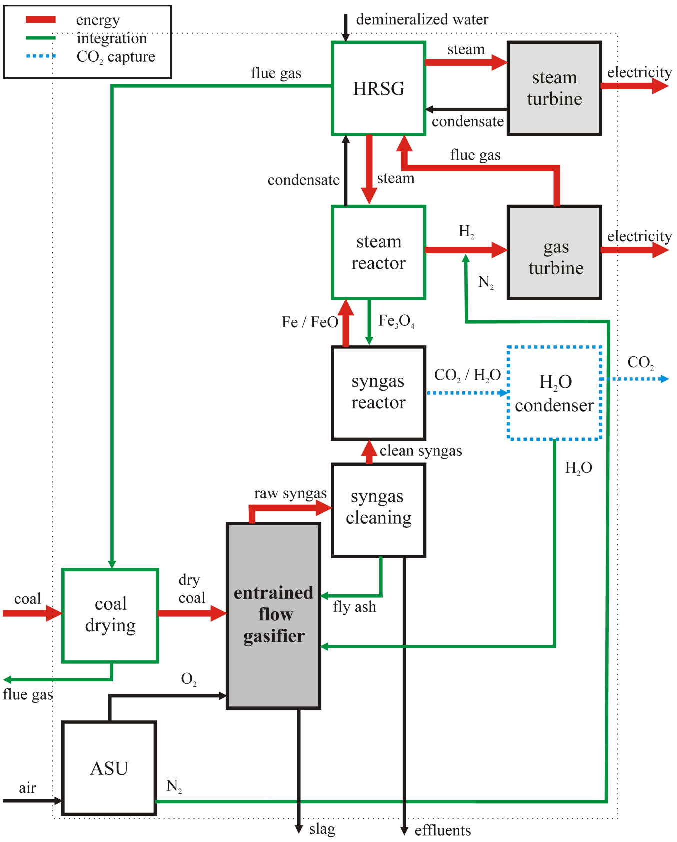

Figure 2: The flowsheet of the coal fuelled oxy-gasification steam chemical looping combustion gas turbine (OG-SCLC-GT) power cycle

generation-derived flue gases. This arises from the fact that biogas contains less impurities such as particulate matter, NO_x, and SO_2. Further, biogas and biogas-derived post-processing gases are more enriched in CO_2 than air-fuel combustion-derived flue gases. This CO_2-enrichment provides an opportunity for low cost CO_2 capture due to the increased driving forces in CO_2 separation processes. Furthermore, by using a WGS reactor the syngas is shifted to a CO_2-H_2 mixture from which the separation of CO_2 is technologically simpler than that from e.g. CO_2-H_2 or CO_2-CH_4 mixtures. The aim of the investigated decarbonized oxy-reforming fuel cell (OR-FC) power cycle is to generate fully decarbonized power via a syngas route from renewable biogas. This concept relies on oxy-fuel CH_4 splitting into CO and H_2. CO is then oxidized to CO_2 providing energy for the endothermic CH_4 as well as H_2O splitting. H_2 is further separated and used as a fuel for electrically efficient fuel cells [10]. Since steam methane reforming is thermodynamically an equilibrium reaction a resulting unreacted CO/H_2O mixture can be either recycled back into an oxy-reforming reactor [11] or shifted to CO_2/H_2 by a selective catalytic water-gas shift (WGS) reaction. While the present study analyzes only the latter option, a combination of the two approaches is also feasible, as well as WGS equilibria can be shifted by separating one of the two products of the WGS reaction, i.e. H_2 or CO_2. Biogas includes completely oxidized carbon in the form of CO_2 and unoxidized carbon in the form of CH_4. The objective of the current OR-FC process is to split CH_4 by means of oxy-reforming into free H_2 and simultaneously completely oxidize the remaining CO to CO_2. In order to avoid any dilution of reaction products with nitrogen, pure oxygen is supplied as an oxidizer. Therefore, the fully decarbonized OR-FC process seeks improved conditions for separating a CO_2-H_2 mixture at the expense of using oxygen as an oxidizer. Oxygen generation can be beneficially integrated in a closed mass and energy recirculation loop with a fuel cell unit and three reactors of the OR-FC cycle. Further, the addition of H_2O is beneficially another source of H_2 as well as oxygen thus reducing the consumption of pure oxygen and increasing the yield of H_2 in product gases. By ensuring strict molar C/O ratio in substrates equal to 0.5 the overall reaction of the OR-FC process can proceed according to the following overall reaction:

$$\begin{aligned} &CH_4 + \frac{1}{1.8}CO_2 + nO_2 + 2(1-n)H_2O \leftrightarrow \\ &\frac{2.8}{1.8}CO_2 + 2(2-n)H_2 \end{aligned} \tag{4}$$

Autothermal operation of the OR-FC process can be achieved when n = 0.3412 [11] and hence eq. (4) becomes:

$$\begin{aligned} &CH_4 + 0.5556CO_2 + 0.3412O_2 + 1.3176H_2O \leftrightarrow \\ &1.5556CO_2 + 3.3176H_2 \end{aligned}$$

$$\triangle H^{298} = 0kJ/mol \tag{5}$$

From eq. (5) it can be calculated that from biogas comprising e.g. 64.29 %v CH_4 and 35.71 %v CO_2 (i.e. CH_4:CO_2 = 1.8) with the addition of oxygen and H_2O in quantities satisfying C/O = 0.5 and C/H = 0.2344 one can obtain by autothermal conversion a mixture comprising 68.1 %v H_2 and 31.9 %v CO_2. Consequently, by utilizing the OR-FC process H_2 can be autothermally produced from renewable biogas with a yield of 3.3176 H_2:CH_4 (molar basis) and the remaining gas beneficially includes almost solely CO_2, i.e. is ready for compression and sequestration. The separation of this CO_2-H_2 mixture is technologically simpler than the separation of CO_2-N_2 or CO_2-CH_4 mixtures. However, the overall reaction given in eq. (5) is reversible and hence the product gases from the OR-FC process include some CO and H_2O, since process equilibria are dominated by WGS chemistry. In order to force shifting of CO/H_2O to CO_2/H_2 two selective WGS reactors are involved. Accordingly, a high temperature WGS (HT-WGS) reactor and a low temperature WGS (LT-WGS) reactor are sequentially set up after the oxy-reforming reactor.

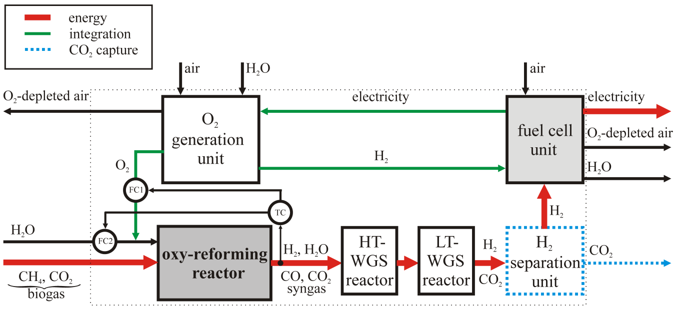

Figure 3: The flowsheet of the OR-FC power cycle. Notation: FC - flow controller, TC - temperature controller

The flowsheet of the OR-FC cycle is presented in Figure 3. Accordingly, after the oxy-reforming reactor the reaction mixture still includes some quantities of CO and H_2O which need to be shifted to CO_2 and H_2. This is accomplished by the inclusion of two WGS reactors. The OR-FC process involves two elementary reversible chemical reactions, i.e. steam methane reforming (SMR) and WGS. The SMR reaction is strongly endothermic and produces moles thus, according to Le Chatelier's principle it is favored at high temperatures and low pressures. High temperatures are thus practiced in industrial conditions. However, in terms of pressure it must be noted that the SMR reaction requires catalysts, which are better utilized under high-pressure conditions. Therefore, the oxy-reforming reactions are usually conducted under high-pressure [12]. In contrast, the WGS reaction is thermodynamically favored by low temperatures. However, to prevent a reduction in the reaction rate, temperatures must decrease gradually, i.e. by passing process gases from the oxy-reforming reactor through the HT-WGS reactor to the LT-WGS reactor. Further, according to the OR-FC process, the separated H_2 is sent to a fuel cell unit. The oxy-reforming and WGS reactors can operate under high-pressure, which is beneficial for H_2/CO_2 separations by increasing their separation driving forces. Besides, high-pressure operation enables improvements in the utilization of catalysts by increasing the amount of fuel gas flow per unit amount of catalysts per unit time. Finally, the fuel cell unit transforms H_2 into electricity with high fuel-to-electricity efficiency. An O_2 generation unit (OGU) can utilize either membrane air separation or H_2O electrolysis operated on electricity generated in the fuel cell stack. In the latter case, H_2 generated by electrolysis is fed to the fuel cell. The whole OR-FC process can thus be beneficially integrated by means of energy and mass recirculation loops. The thinking underlying the proposed concept of biogas conversion via the OR-FC cycle with negative net CO_2 emissions can also be useful in other similar biogas conversion cycles.

3. Conclusions

The paper analyzed low-carbon power generation concepts. Three promising low-carbon power cycles were examined with an emphasis put on the feasibility of CO_2 capture and integration opportunities. It was shown that high CO_2 capture rates were feasible from pressurized CO_2-enriched streams comprising either water or hydrogen, thus necessitating oxy-fuel power cycles. Opportunities for mass and energy integration were found to be increased in systems involving closed mass and energy recirculation loops. The discussions also emphasized that low-carbon power cycles could achieve minimized exergy losses by applying more reversible energy conversion processes. The first low-carbon power cycle under study, the MSG-GT cycle, offered the possibility of avoiding problems encountered in the two current large-scale biomass-based power technologies: (i) one-step direct combustion (low efficiency) and (ii) co-combustion with coal usually in centralized power plants (need to transport biomass over long distances). The MSG-GT cycle investigated involved biomass pre-treatment via drying and torrefaction followed by biomass gasification to syngas and high efficiency syngas combustion. It was emphasized that such a multi-step cycle led to more reversible fuel processing than achieved in one-step combustion. Therefore, in accordance with the principle of entropy equipartition, entropy was generated more uniformly in all sub-units, which means less exergy destruction and thus increased cycle efficiency. The second low-carbon cycle investigated, the OG-SCLC-GT cycle, benefited from both improved cycle efficiency and the feasibility of extremely high CO_2 capture rate. In the OG-SCLC-GT cycle coal was dried, gasified in the oxygen-blown gasifier and the resulting syngas was fed to the syngas CLC reactor that produced a CO_2/H_2O mixture. Further, the reduced oxygen carrier was used in the steam reactor to produce hydrogen, which was sent to the combined cycle gas turbine. The OG-SCLC-GT cycle was superior in the use of advanced mass and energy integration via the inclusion of several closed recirculation loops and due to the extremely large CO_2 capture rate. The third decarbonized power cycle, the OR-FC cycle, offered to produce hydrogen from renewable biogas with a maximal yield of ca. 3.3 H_2:CH_4 on a molar basis in autothermal conditions. Through capturing CO_2 the OR-FC power cycle achieved negative net CO_2 emissions. The OR-FC power cycle involved the oxidative reforming of biogas to syngas followed by a high-temperature shift reaction and finally a low-temperature shift reaction was performed to produce H_2. The OR-FC process was integrated with CO_2 capture, the fuel cell and the oxygen generator. The main advantage of the OR-FC process over other existing decarbonized processes included H_2 separation from highly concentrated CO_2-H_2 mixtures while the remaining CO_2 could be compressed and sequestered. Pure hydrogen was converted into electricity in the efficient fuel cell. Also energy and mass integration was assured by involving closed recirculation loops.

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Nomenclature

CCS	carbon capture and sequestration
C/H	C to H ratio in a mixture (molar basis)
CLC	chemical looping combustion
C/O	C to O ratio in a mixture (molar basis)
GT	gas turbine
HT-WGS	high-temperature water gas shift
IGCC	integrated gasification combined cycle
LCE	Low-Carbon Economy
LT-WGS	low-temperature water gas shift
MSG-GT	multi-step gasification gas turbine
n	stoichiometric coefficient in eq. (4)
OG-SC- -LC-GT	oxy-gasification steam chemical looping combustion gas turbine
OR-FC	oxy-reforming fuel cell
SCLC	steam chemical looping combustion
SMR	steam methane reforming
WGS	water gas shift

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