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Article

Energy Efficient Design of Power-to-Gas Generation Systems for Circular Economy

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Abstract: Generous subsidies and "technological openness" cannot fulfill European goals of affordability, environmental compatibility, and security of supply as recent blackouts and rising grid fees show. A clear structured definition of development targets based on TRL (Technological Readiness Level) is needed as used in aerospace industry. Affordability and security of supply provide the guidelines for a CO_2 emission free system design. Energy transition will only work with a sufficient storable thermodynamic potential that guarantees power generation at any time with a cost-effective energy storage technology. While H_2 suffers from its low volumetric energy density and high cost a Closed Carbon Cycle (CCC) based on CH_4 and C_2H_4 delivers an energy density up to more than 16 times higher than H_2 with increasing carbon content. High thermodynamic system integration, including heat pumps and electrochemical O_2 removal during the hydrocarbon-forming reaction, increases efficiency. Electrolyzers and CH_4/C_2H_4 forming reactors can be integrated in a steam generator design to optimize internal heat transfer and allow variable pressures. The conversion efficiency is estimated at up to 90% and a reconversion efficiency like pump storage hydro power plants. The concept fits very well with the current infrastructure of natural gas. It can supply industry with renewable hydrocarbons and use plastic waste for recovery. "Green fuels" can be delivered by using CO_2 from bio-energy utilization. H_2 can be additionally generated by reforming like today with natural gas. Conversion efficiency, electricity price, full load operation hours, and investment cost mainly influence the gas price.

Keywords: System Specification; Efficiency Improvement; Design Principles; System Integration of Circular Economy; Economic Feasibility

1. Introduction

The conception of the intended energy transition based on the Paris agreements hardly fulfills the requirements of the most holistic view possible, which a sustainable industrial society requires, but focuses solely on the reduction of CO_2 emissions. It gives the impression that the rise in the price of CO_2 certificates—as the so-called acceleration tool of any improvement—is the target function of any action rather than a sustainable technologically advanced society itself. However, the real task should be to present an efficient, feasible, resilient and sustainable system of future energy supply, which has not been done so far. Subsumed under the term "technology openness," which seems reasonable at first, a collection of different individual solutions has emerged over the course of time, some of which make perfect sense without it being clear how a functioning overall system is to be created from them. In view of the goal to be achieved, it is necessary to identify solutions for a functioning renewable energy system that simultaneously enable a sustainable supply of raw materials in a circular economy by means of closed

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carbon cycles (CCC). Closed cycles make it possible to utilize the advantages of hydrocarbons as an energy source and as an industrial raw material without releasing CO_2 into the atmosphere, as is the case in nature. The results of our own study on the energy transition analyzing previous concepts showed that they do not lead to cost efficiency or security of supply [1]. The results presented here explain the solution with CCC. The underlying thermodynamic analysis opens up new ways to the necessary improvement in the efficiency of gas generation and enables the use of ethene (C_2H_4) as a storage gas. The existing gas storage facilities are thus sufficient to enable a secure renewable primary energy supply at today's level and at the same time the transport capacity of the gas network is significantly increased. The possibility of the associated increase of renewable electricity generation does not require a compulsory, uneconomical increase in energy efficiency of the entire infrastructure, which is significantly more expensive than maintaining current primary energy demand. This analysis explains the system specification, synthetic gas generation and the basics of efficiency improvement, design principles of gas generation, as well as system integration into a circular economy and its economic viability. The concepts and study results of our own work in recent years presented here in a compact form can be explored in more detail in the citations of our own papers provided. The development goals must be defined based on the actual TRL of the system and components. Their different development stages range from TRL 2/3 (basic research/proof of concept) to TRL 9 (system testing, market launch, and operation) [2].

2. System Requirements for the Use of Renewable Energy for Power Supply

Reliability of supply is the first rule for planning any energy system. In the field of renewable energy was recognized at an early stage in the 1950s that any renewable energy source need storage for a sufficient reliability. The basic considerations of a hydrogen economy for the solution of a future energy shortage assumed solar primary energy, which inevitably involved the storage of large amounts of energy [3]. In contrast, Czisch assumed that a widespread electrical grid with correspondingly widespread distribution of wind feed-in would be sufficient for a secure supply and would require only small backup power [4]. From the latter, the thesis developed that the feed-in of wind and solar energy complement each other well on average and that at most two-week dark doldrums are to be expected, and thus the storage problem is only of secondary importance. Therefore, the storage problem was not deeply investigated compared to planning of wind parks and different solar installations and a comparable small battery capacity was expected to be sufficient.

But the reality showed that that low wind power production in winter over a longer time could not cover the demand in a longer period than only two weeks and a considerable solar power was usually not available at this time and the assumed battery capacity was far too low. The only reason that a blackout could be avoided was that fossil and nuclear power plants were still at operation and not scrapped. Obviously the thermodynamic background of save grid operation need to be explained by using **Figure 1**.

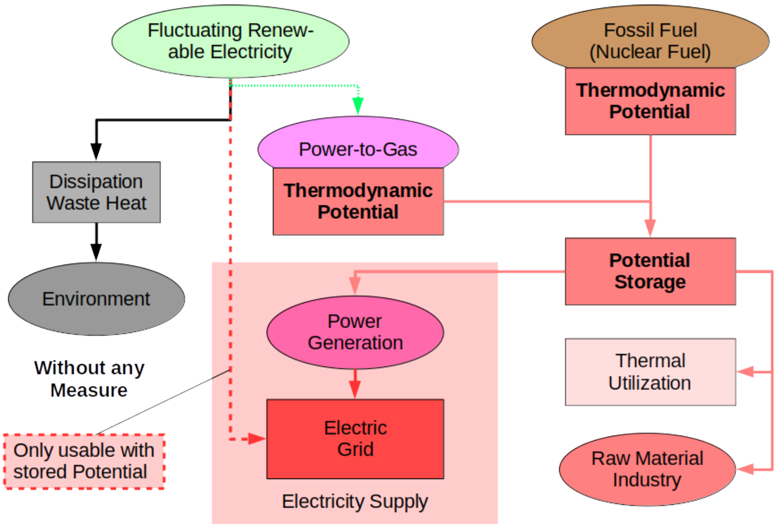


Figure 1. System Requirements for the Operation of Reliable Power Grids.

The just on demand production of power needs an always available thermodynamic potential as e.g., represented by any fuel or the height differences for water reservoirs. Without this, fluctuating renewable electricity supplied to the grid would generally be dissipated as heat immediately. Therefore, a sufficient portion must first be converted into thermodynamic potential. This allows controllable power plants to compensate for the difference between renewable supply and grid demand, stabilizing the grid. Additionally, converting renewable electricity to thermodynamic potential is necessary to store excess supply for periods when renewable electricity is insufficient. In principle, batteries or synthetic burnable gases can be used as an energy storage. The choice of technology depends, besides cost, mainly on the amount of potential that needs to be stored.

An analysis of the storage demand based on the different monthly electricity generated by wind and solar generators from 2011 to 2019 is presented in the own study [5]. The monthly values are related to the annual production of the wind and solar generators to get a comparable set of Figures. However, it is difficult to find an exact value for the annual storage demand but a first estimation can be made based on these Figures. **Figure 2** shows the results. On the left side is the maximum monthly demand shown over the seasons compared with the minimum monthly supply of solar and wind. Parameter is the variation of the share of solar production. The right side of the Figure shows the annual difference between maximal demand and minimum supply over the share of solar supply. These values of deviation shows the minimum demand of stored energy between about 20 to 30% of the annual demand in the relevant scenario. It shows that an increasing wind supply increases the storage demand. The minimum storage demand can thus be estimated between about 20 and 30% of the annual demand.

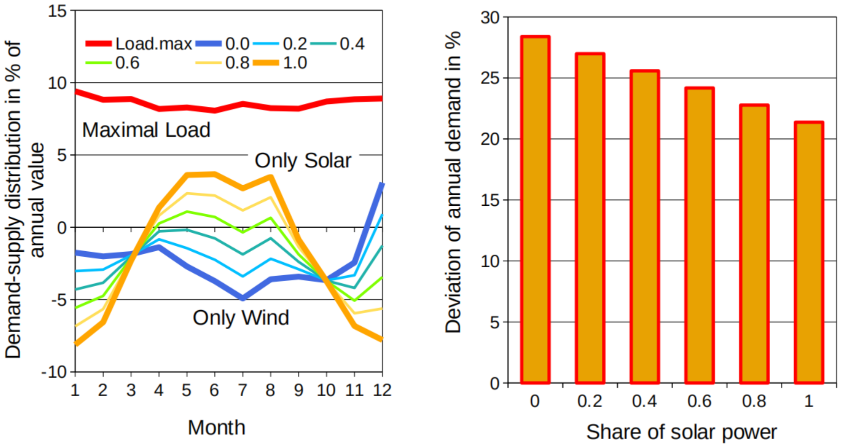


Figure 2. The Share of Solar Generation and Its Influence on Storage Design.

However. the estimation of **Figure 2** gives a first impression based on operation data there is another important source to see the storage demand. This is a view on the existing storage facilities to guarantee a stable natural gas supply in Europe as given in **Table 1** [6,7].

Table 1. Actual Existing Storage Capacity of Natural Gas in Europe in 2021 [6,7].

Country	**Annual Consumption in TWh/a**	**Storage Capacity in TWh**	**Fulfillment of Demand in %**
Germany	908.3	250.6	27.6
Italy	726.1	195.2	26.9
Netherlands	350.6	142.4	40.6
France	430.4	133.6	31.0
Austria	89.9	96.7	107.5
Hungary	108.1	69.7	64.5
Czech Republic	90.7	44.2	48.7
Poland	212.0	37.5	17.7
Slovakia	52.9	37.1	70.1
Denmark	22.5	9.8	43.6
Portugal	57.8	4.0	6.9
Total Europe	3049.4	1020.8	33.5
Germany H_2-1	908.3	58.4	6.4
Germany H_2-2	211.5	58.4	27.6

The supply of natural gas via pipelines or the sea is far more reliable than that of renewable energy, which means that at least a third of the annual requirement needs to be stored. This makes batteries uneconomical. According to **Table 1**, H_2 is also unsuitable as a strategic energy reserve, as fulfillment of demand would fall to 6.4% if consumption remained constant (scenario H_2-1) or only 211.5 TWh could be consumed annually if fulfillment of demand remained the same. This raises the question of suitable storage gases, whereby in addition to the natural gas (CH_4) used today and the H_2 currently favored by politicians, C_2H_4 is also an option. An initial impression can be gained by plotting the density against the system pressure. As **Figure 3** shows. The original values are shown on the left and the density ratio of CH_4 and C_2H_4 in relation to H_2 is plotted on the right to illustrate the differences [8,9].

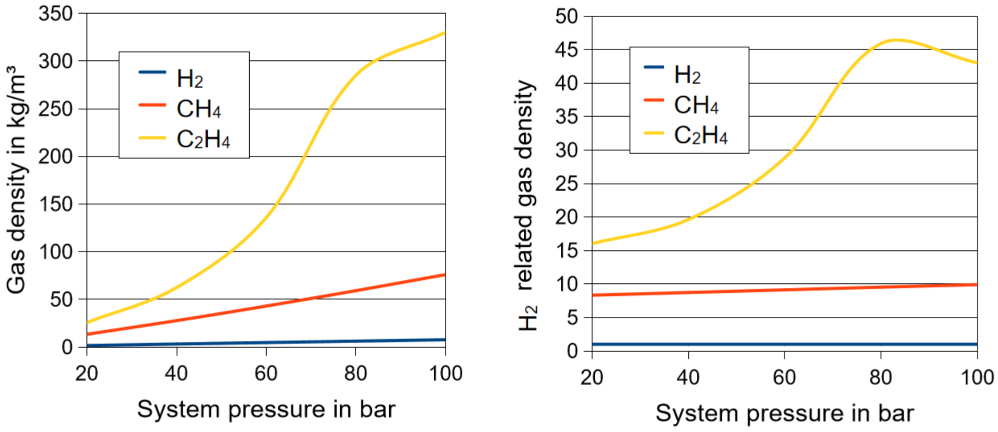
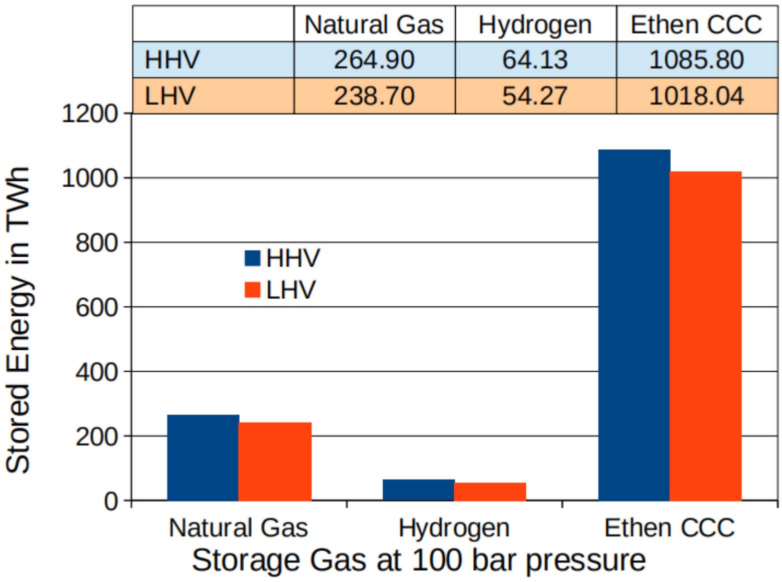


Figure 3. Density of Possible Storage Gases Above Their Pressure [8,9].

The density of the possible storage gases increases very strong with the increase of the carbon content of the storage gas. Consequently, the use of C_2H_4 obviously allows a significantly larger utilization of the storage volume than it is possible with natural gas and thus a significant increase of the storage capacity is principally achievable without additional costs. However, mass related LHV of H_2 is clearly higher than these of CH_4 and C_2H_4 but not that much to change this picture as **Figure 4** shows [7].



	Natural Gas	Hydrogen	Ethen CCC
HHV	264.90	64.13	1085.80
LHV	238.70	54.27	1018.04

Figure 4. Energy Utilization of the Existing German Gas Storage Volume in TWh [7].

The suitability of storage gases generated with power-to-gas (PtG) essentially depends on the emissions resulting from the intended use. For the use described here in the context of a carbon cycle economy, the question of CO_2 emission is basically irrelevant because CO_2 is an essential raw material of the overall process and thus must not leave the process. Nevertheless, the conversion of the storage gas into H_2 without CO_2 emissions needs to be considered, because in all open processes H_2 will always be used when a storage gas is needed. The existing infrastructure also suggests synthetic CH_4 and C_2H_4 as substitutes for natural gas. The key criterion for suitability as a storage gas is its energy density as a function of pressure, as shown in **Figure 4**. This clearly shows that the ideas of a hydrogen economy based on a H_2 used as a countrywide strategic energy storage will fail dramatically, but H_2 production for local use supplied by a carbon cycle economy as natural gas is used for H_2 production today. High storage and transportation costs of H_2 could be avoided. The following two paragraphs describe the thermodynamic proof of concept and thus the principal feasibility of proposed technical solutions (TRL 2/3).

3. Principles of Synthetic Generation of Storage Gases

The own investigation of possible combinations of process chains to produce CH_4, and C_2H_4, based on reversible structures showed that both species can be produced in similar ways [10]. These principal thermodynamic possible reactions were analyzed by a systematic approach to identify these reaction steps and the impact of their components and structure on energy efficiency of real systems as shown in **Figure 5**.

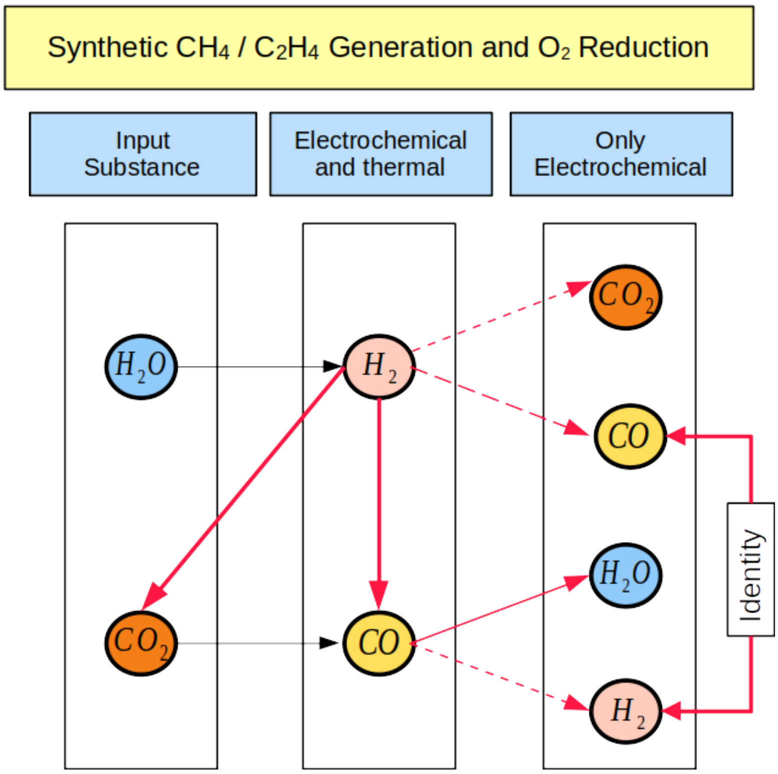


Figure 5. Structural Investigation of Possible Reversible Process Combinations.

The status of the O_2 separation within the possible combinations of the process design can be analyzed by using three columns. The first column represents the input molecules H_2O and CO_2. The second column represents the possible reactions of H_2 and CO generated by electrolysis from the input molecules and can be performed by thermal reactions. The third column represents the process options of possible electrochemical driven reactions instead of the thermal process. The column in the middle in combination with the first column shows that we can only combine H_2 either with CO_2 or CO to generate CH_4/C_2H_4 in a thermal process. The combination of the middle column with the third column allows the combinations of H_2 with CO and CO_2 and of CO with H_2O.

The electrolysis of H_2O and CO_2 allows thus principally 5 basic reactions that are possible for methanation and also for direct C_2H_4 generation. The work demand of the methanation and C_2H_4 formation is called "electrochemical" for simplicity however the reversible thermodynamic analysis can only say what minimum work is needed and not its type. The reversible thermodynamics can only express the work and heat demand or its need for a certain process but not the technical specification of the type of work.

Furthermore, the use of an oxygen conducting membrane for the O_2 removal is assumed however a detailed investigation is neglected at this stage. It is also currently useless to discuss the exact gas quality because the main topic is not the production of a pure CH_4 or C_2H_4 but of an artificial fuel gas that is usable as energy storage and basic chemical. Thus, it may have a tolerable content of CO_2, CO, and H_2 as well. Anyway, this has to be mentioned here to show the limitations of this investigation and to explain further research demand.

The reactions studied above are numbered and summarized in **Table 2**, including the amount of O_2 removed and the method used. The similarity of the reaction equations reflects with the methodology and the amount of O_2 removal in the last reaction step. The first two reactions are thermal driven while the last three depend on external work supply. The similarity of the reactions and associated processes of CH_4 and C_2H_4 forming can be easily identified. The further explanations for CH_4 are thus also valid for C_2H_4 without further notice.

Table 2. Overview of Identified Reversible Reactions for CH_4 and C_2H_4 Forming.

No	CH_4	C_2H_4	O_2 Removal	
1	$4H_2 + CO_2 = CH_4 + 2H_2O$	$6H_2 + 2CO_2 = C_2H_4 + 4H_2O$	via H_2O	thermal
2	$3H_2 + CO = CH_4 + H_2O$	$4H_2 + 2CO = C_2H_4 + 2H_2O$	via H_2O	
3	$2H_2 + CO = CH_4 + 1/2O_2$	$2H_2 + 2CO = C_2H_4 + O_2$	via O^{2-} Conduction	electro-chemical
4	$2H_2 + CO_2 = CH_4 + O_2$	$2H_2 + 2CO_2 = C_2H_4 + 2O_2$	via O^{2-} Conduction	
5	$2H_2O + CO = CH_4 + 3/2O_2$	$2H_2O + 2CO = C_2H_4 + 2O_2$	via O^{2-} Conduction	

The following considerations can now be referred to this Table and a connection between the reaction equation and the O_2 removal is easily understood. The considerations are focused on the following three major aspects:

(1) Discussion of the dependencies of the different investigated systems,
(2) Efficiency limitations, and
(3) Analysis of possible improvement and necessary research.

The use of reversible process chains allows following the own method an analysis of improvements with real process structures [11]. The reversible process is the border case of any real process thus reversible structures are also the best possible real process structures. The only limitation is the availability of components and that only approximations are the possible solution. **Table 2** shows that there are only two process structures needed the thermal methanation and the electrochemical. The process description is given in **Figures 6** and **7**. In both cases the reverse reaction of CH_4 combustion is the starting point.

The removal path of O_2 is the guideline of the process description. The total mols to be removed are 2. The first step is to reduce the O_2 of the inlet H_2O and CO_2 by electrolysis. But this delivers only a reduction by 1 ½ mol O_2. Therefore, an additional mol H_2 is needed to remove this ½ mol O_2 by forming 1 mol H_2O in the methanation reactor.

The electrochemical methanation needs in principle the same electrolysis steps for H_2O and CO_2 as the thermal methanation. The only difference is the extraction of O_2 from the methanation reactor. Following the example of **Figure 6** again in **Figure 7** ½ mol O_2 needs to be removed as well. An O^{2-} conducting membrane allows principally a separation of O_2 in situ. However further investigations and research are needed but this process seems to be flexible because the same design should be usable for all described reactions No 3 to 5 (**Table 2**).

However, the investigation of storage capacity of synthetic gases showed a great opportunity for CH_4 and C_2H_4 but there is still the problem that the efficiency of all cycles based on synthetic gases have a too low cycle efficiency as different sources as shown in the work of Beck et al. [12]. Therefore, the improvement of cycle efficiency is absolutely needed for the successful use of synthetic gases.

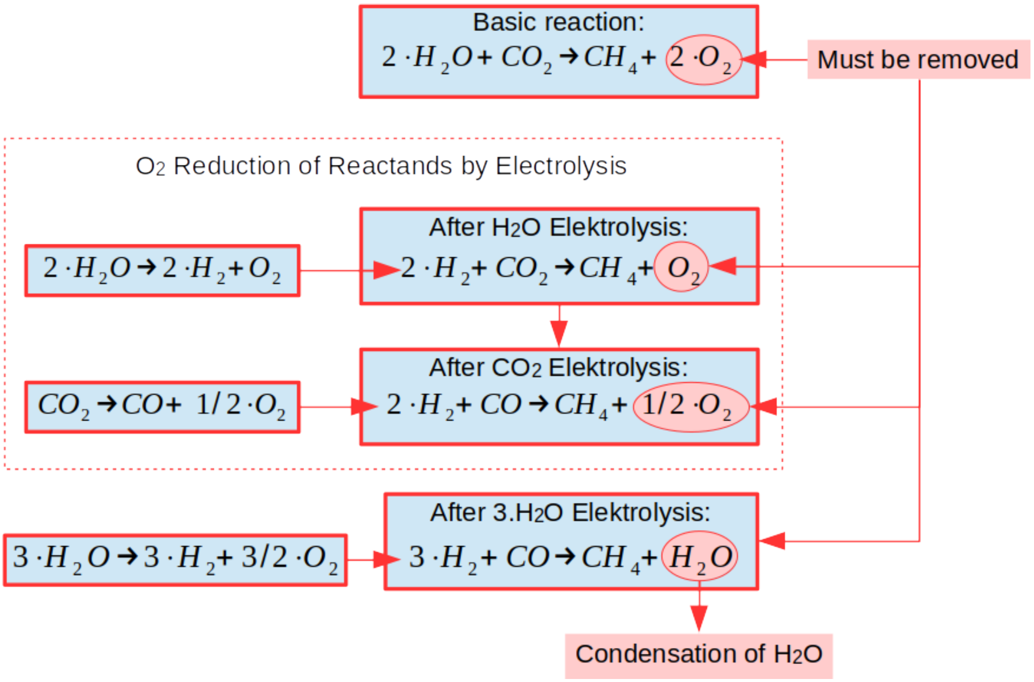


Figure 6. Process Description of the Thermal Methanation.

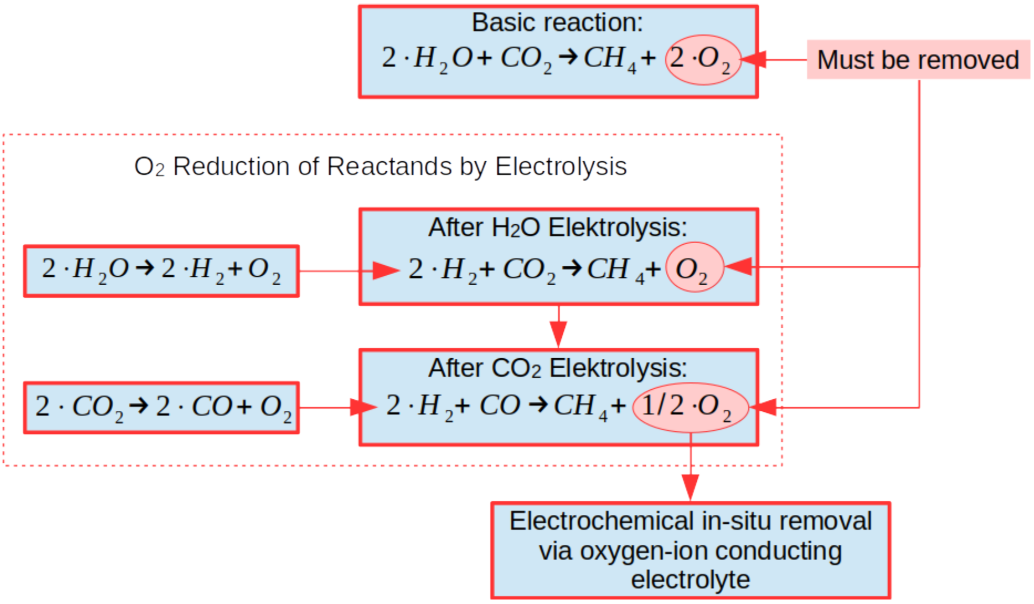


Figure 7. Process Description of the Electrochemical Methanation.

4. Improving Process Efficiency

However, electrolysis and electrochemical fuel utilization especially with H_2 are known since the 19th century they are not seen connected in a cycle. The H_2O content in the flue gas of boilers and other combustion processes was considered by the description of fuel quality by HHV (Higher Heating Value) and LHV (Lower Heating Value) delivering a sufficient solution for a fossil world. But a renewable hydrogen economy has other rules. It depends principally on a systematic connection of fuel generation from renewable electricity and its independent utilization from the fluctuation of the renewable electricity production. The above delivered process description of storage gases already shows the importance of the H_2O electrolysis for the synthetic storage gas generation. The use of reversible cycles is an efficient method to understand the basic principles of processes to be investigated. The here needed reversible cycle is shown in **Figure 8**.

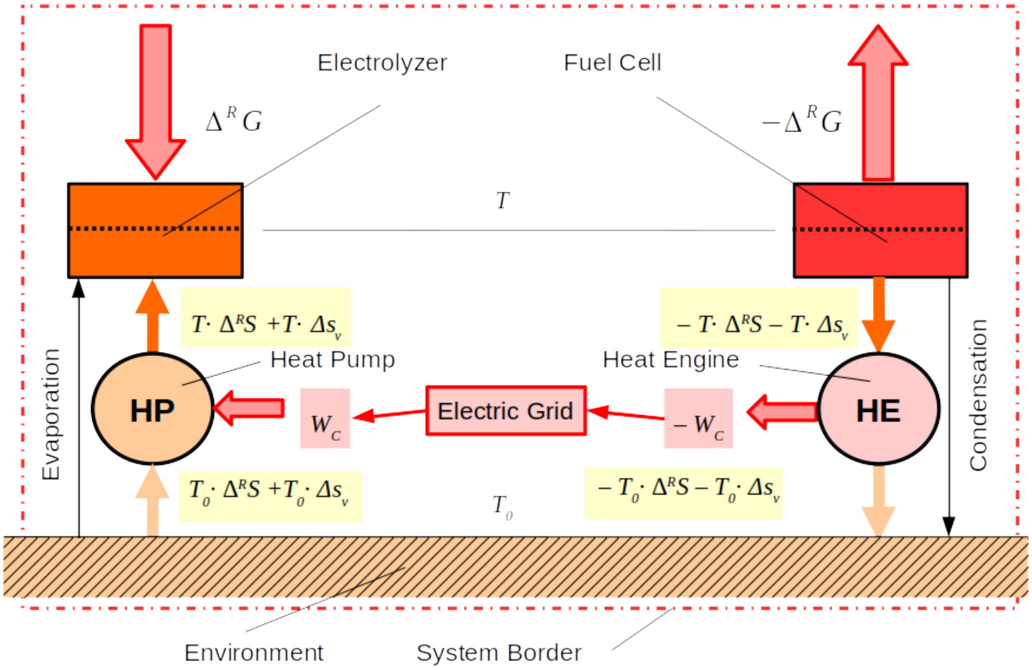


Figure 8. Reversible Reference Process for the H_2 Utilization Cycle.

The reference process consists of a reversible H_2 electrolyzer and a reversible H_2 converting fuel cell, both with a constant operation temperature T, built in an environment with the temperature T_0. The produced H_2O is released to the environment and reused for feeding the electrolyzer and a reversible water processing is assumed. The electric grid, the H_2 transport and storage between electrolyzer and fuel cell are also assumed as reversible. These assumptions allow to only consider the reversible operation of electrolyzer and fuel cell. Therefore, only the entropy change by evaporation Δs_{ev} and the reaction entropy $\Delta^R S$ has to be considered. The condenser releases Δs_{ev} at the temperature T to the ambient state T_0 by producing the reversible work—$w_{trev.ev}$ as the Carnot work—w_C. The energy balance delivers for the condenser of the fuel cell considering only the evaporation heat:

$$0 = T \cdot \Delta s_{ev} - T_0 \cdot \Delta s_{ev} - w_{trev.ev} \tag{1}$$

and for the evaporator of the electrolyzer:

$$0 = T_0 \cdot \Delta s_{ev} + w_{trev.ev} - T \cdot \Delta s_{ev} \tag{2}$$

There is no change of entropy in this process and the amount of transferred heat is only defined by its temperatures, because heat engines, heat pumps, and heat transfer have been assumed as reversible in this process. The already above addressed high losses of actual electrolyzers are caused by the fact that there is no recovery option of the condensation heat of fuel cells seen as possible. But on the other side the exergy destruction by dissipating electric work to evaporate the incoming liquid H_2O leads to an efficiency loss of 15% and totally to 17% including reaction entropy demand. However, every experiment shows the so called “Thermoneutral Voltage” representing the total energy demand of an ideal electrolyzer supplied with liquid H_2O [13]. Anyway, this heat supply by dissipation is not an electrochemical reaction at all that needs to be performed in the electrolysis cells. From a point of system thermodynamics it is necessary to replace electricity as an energy source of evaporation by any method of heat recovery or any other low grade heat source to improve the electrolyzer efficiency. **Figure 9** summarizes possible options.

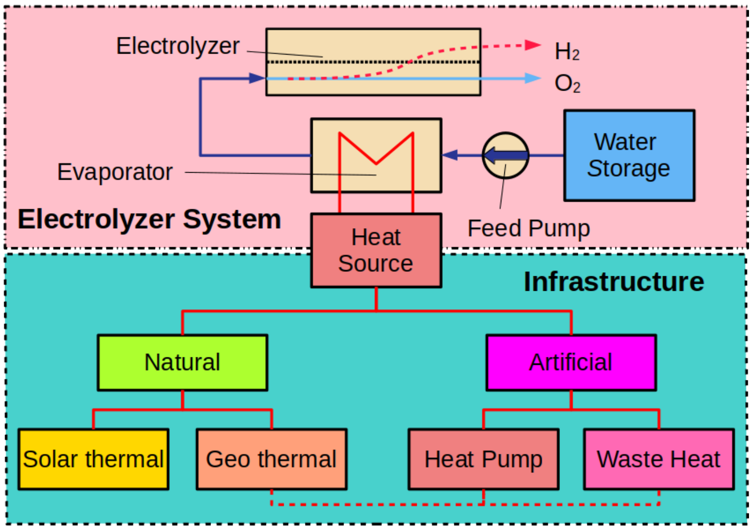


Figure 9. Overview of Possible Heat Sources for Evaporation in Electrolyzer Systems.

The used heat sources can be natural as solar or geothermal heat or artificial as waste heat from different industrial or other heat delivering processes. The use of heat pumps allows to use also low temperature heat below the evaporation temperature. This consideration shows that an intelligent system integration could clearly improve the process efficiency of electrolysis to an order of 90%. The reported efficiency of methanation is currently clearly lower than the data from electrolysis [12,14]. Referring to the potential of CH_4 and C_2H_4 as storage gases, it is very important to investigate the further improvement possibilities based on the scientific fundamentals.

The improvement possibilities of CH_4 and C_2H_4 generation systems can be understood by investigating the thermodynamics of the entire possible reactions as first published in previous works where further details can be found in the own studies [10,15]. For the principal understanding, it is sufficient to consider the work demand of the reactions following **Table 2**, as shown in **Figure 10**.

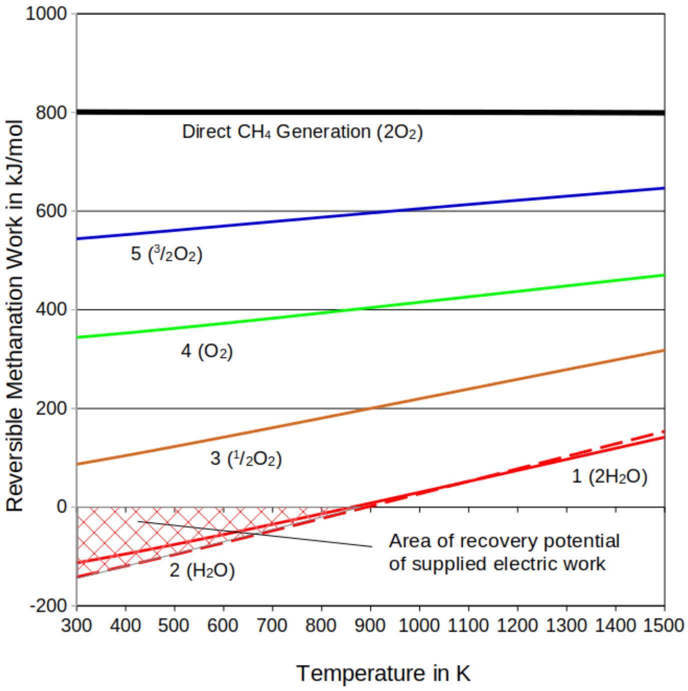


Figure 10. Reversible Work Demand for Methanation Depending on T and the Direct O_2 Removal as Parameter.

The reversible work demand of methanation is plotted versus temperature with the type of O_2 extraction as the parameter. The numbers as shown in **Table 2** identify the type of reaction and the associated O_2 extraction is noted in brackets. All reactions show an increasing work demand with increasing temperature, only the direct reaction of CH_4 generation is not depending on the reaction temperature *T*, as already expected. The two thermal methanation reactions (1,2) deliver below about 900 K reversible work and an increasing work demand with further increasing temperature has to be discharged by an increasing reversible heat due to an only very slightly decreasing of the reaction enthalpy with increasing temperature. The heat management at this temperature is challenging or limited because clearly increasing cost due to the BoP (Balance of Plant) extension for recovery of an increasing heat. The increasing size and cost of BoP with increasing temperatures is a general result for all considered cases of **Table 2** and **Figure 10** respectively due to the increasing heat to be discharged from the system. The general finding of the thermodynamic consideration of the investigated systems shows that the reaction temperature should be chosen as low as possible to allow the design of cost effective CH_4, and C_2H_4 generation systems. These results lead also to the above demanded improvements of the systems.

The current technology is based on the thermal processes, (1,2) in **Table 2**. **Figure 10** shows for both reactions a potential of work recovery at temperatures below about 900 K for CH_4 and C_2H_4 generation as well. The thermal reactions allow thus thermodynamically a supply of the reaction with H^+ ions via a fuel cell injector as shown in **Figure 11**.

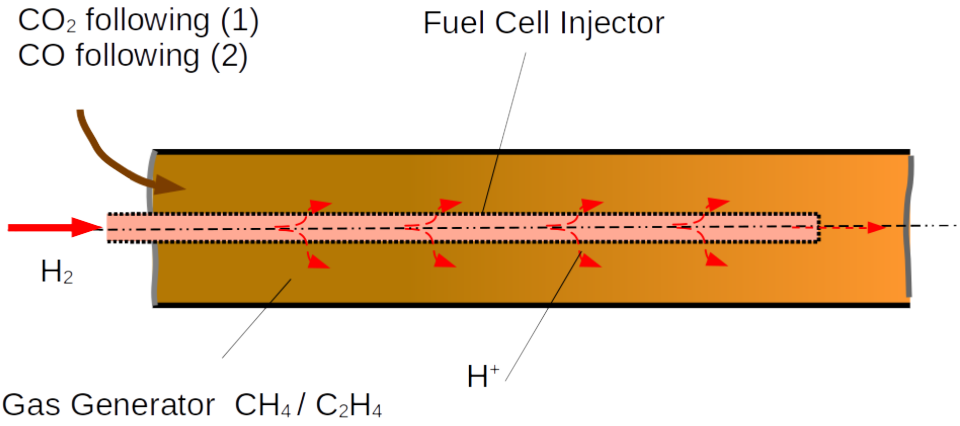


Figure 11. Fuel Cell Injector for Thermal CH_4/C_2H_4 Forming.

Proton conducting electrolytes needed for this application are state of the art. The second possible improvement is the recovery needed for the additional heat supply for the extra H_2 demand in the thermal reaction to remove O_2, as shown in **Figure 12**.

The electrolyzer is supplied by two H_2O sources, one supplied the recovery of the condensation heat and the other by external low grade heat Q_{HP} e.g. via a heat pump. The needed evaporation temperature can be extracted from the condenser via any heat pump system. One solution is to increase the condensation temperature by a steam compressor. The only condition for the design of the heat pump system is to guarantee the evaporator is supplied with a higher temperature than the evaporation temperature.

The electrochemical methanation or the electrochemical C_2H_4 forming generation needs a simultaneous O_2 removal in situ with the reaction itself. The expected benefit of the electrochemical methanation is that the H_2 production can be reduced to its stoichiometric minimum of $2H_2$. Obviously, the design of a mixture of possibly two different catalysts in one reaction chamber and its design could be very challenging and needs further research.

But currently there is the option for an approximate solution by forming electrochemical elements consisting of a small thermal methanation reactor and a seriell connected recovery electrolyzer with an oxygen ion conducting electrolyte. The single modules are seriell connected to form the segmented methanation reactor. This concept is applicable for the electrochemical C_2H_4 forming generation as well. The methanation is now described as a reference for both or similar reactions in **Figure 13**.

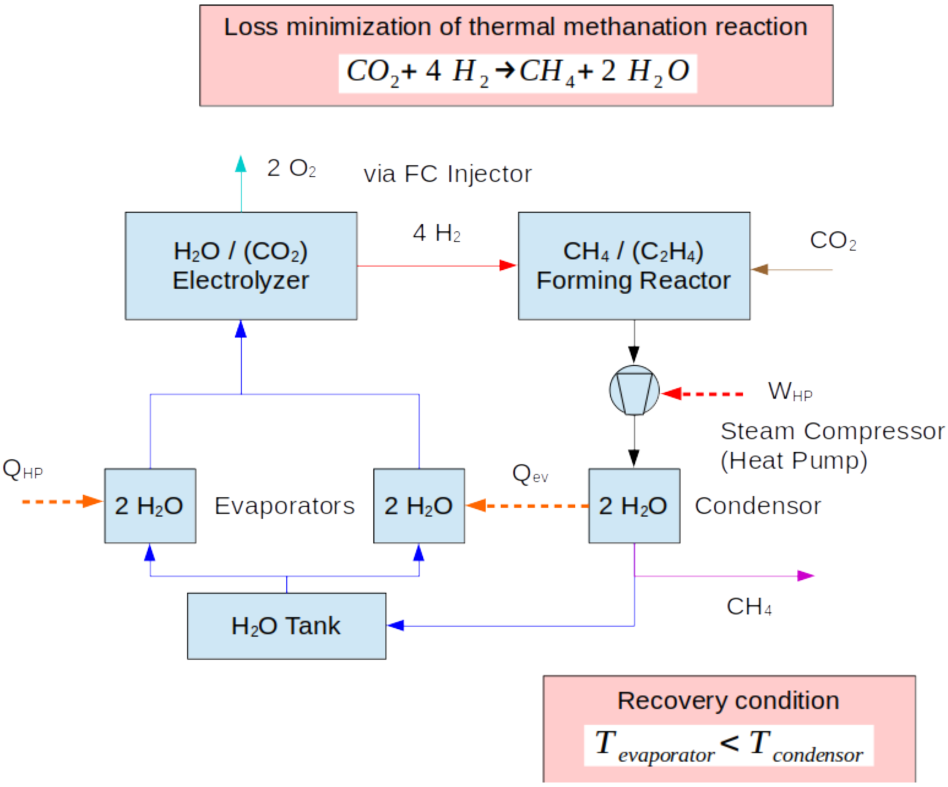


Figure 12. Heat Recovery of Evaporation in Thermal Methanation.

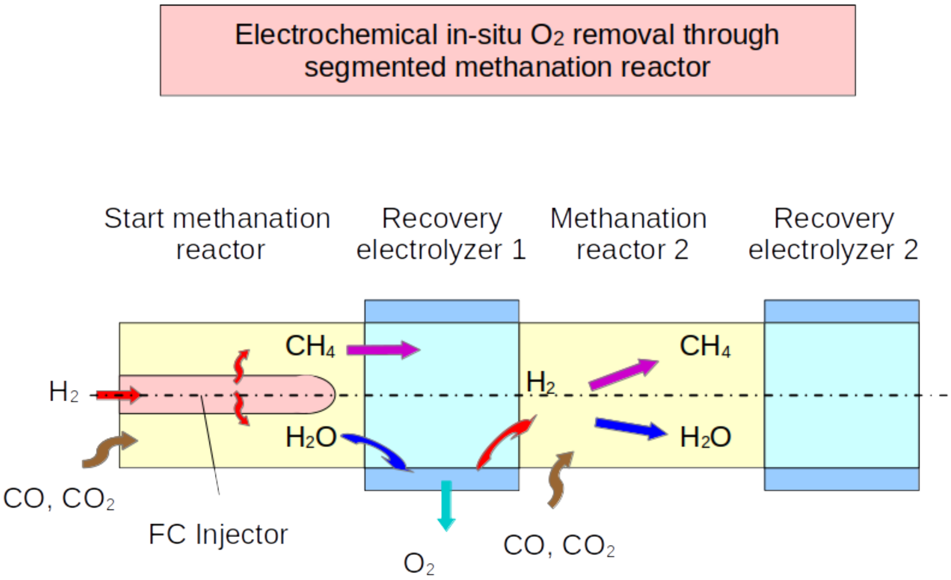


Figure 13. Segmented Electrochemical Methanation Reactor.

The benefit of this assembly is that the H_2 inlet flow of the first module is very close to $2H_2$ because the excess H_2 can be limited to the demand of the first module because the downstream recovery electrolyzer of the module recovers the excess H_2 for reuse in the next module. The benefit of this design is that only a small quantity of excess H_2O is needed and consequently the loss of the extra evaporation heat needed is small. The use of the fuel cell injector and a heat recovery of the product gas is an economic question if the additional investment is paid by the improvement.

5. Design Principles of Basic Reaction Systems

The design structure of the electrolyzer as an essential process component determines the overall concept of gas generation. The electrolyzer design itself is determined by the delivery of steam and is shown in **Figure 14**. However main components of chemical/electrochemical reactors can be only classified as TRL 2/3 but the system itself can be classified as TRL 8/9 because it is nothing else than a modification of existing steam boiler designs as the Figures below show.

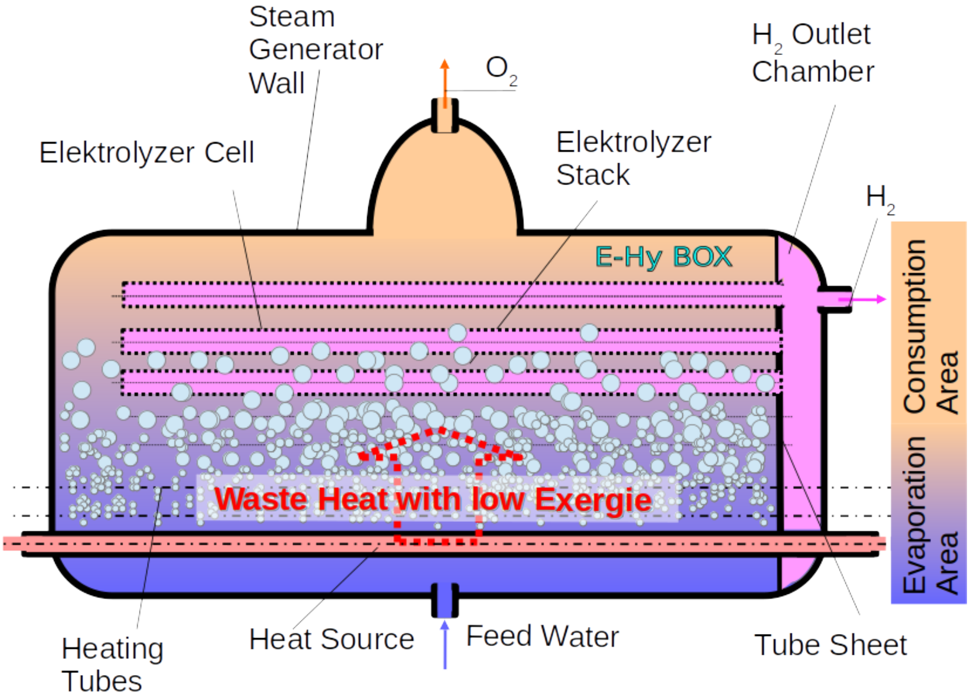


Figure 14. Basic Design of Steam Operated Electrolyzer.

The basic design requirements for the steam generation are similar to long proven boiler concepts and components. These examples are shell boilers and boiler drums of natural circulation boilers. The operation pressure is very flexible for these designs as state of the art natural circulation boilers have an operation pressure up to 180 bar. Thus the operation conditions of the electrolyzer system design are very flexible for any demand. Possible restrictions are thus only caused from the reaction requirements itself.

The design principles of the components inside the steam generator walls are close to shell boilers, the assembly of the electrolyzer modules is very similar to the flue tubes and the assembly of the heating tubes is very similar to that of the flame tube of the shell boiler. The assembly of the electrolyzer modules itself can include cladding tubes to separate the electrolyzer and the boiling water/saturated steam mixture leading to additional pipework and separation compartments [10].

Figure 14 shows the most simple and elegant solution with an open electrode to the steam section. The electrolyzer separates the produced H_2 and O_2. The outlet gas depends on the used electrolyte of the electrolyzer. Proton conducting electrolytes deliver H_2 to the gas outlet chamber and O_2 via the gas outlet dome. O_2- conducting electrolytes change the gas outlets compared to proton conducting electrolytes and the gas outlet chamber delivers O_2 and outlet dome delivers H_2. The heating tubes are supplied by a heat source supplying ideally isothermal heat that may be an exothermic chemical reaction or a phase change as, e.g., condensing steam.

The steam supplied electrolyzer can be easily used as a first stage of a methanation or a C_2H_4 forming reaction because both deliver their exothermic reaction to heat up the steam boiler. Consequently, above the presented electrolyzer system concept can be also used as first stage for methanation or C_2H_4 forming reaction as well. Again, the methanation is used for the description of the design. The different possible methanation reactions following **Table 2** need a clearly more complex BoP than electrolyzers due to the need of heat recovery and CH_4 delivery. **Figure 15** shows the principal BoP of methanation with the reactant CO_2 following the reaction (1) in **Table 2**.

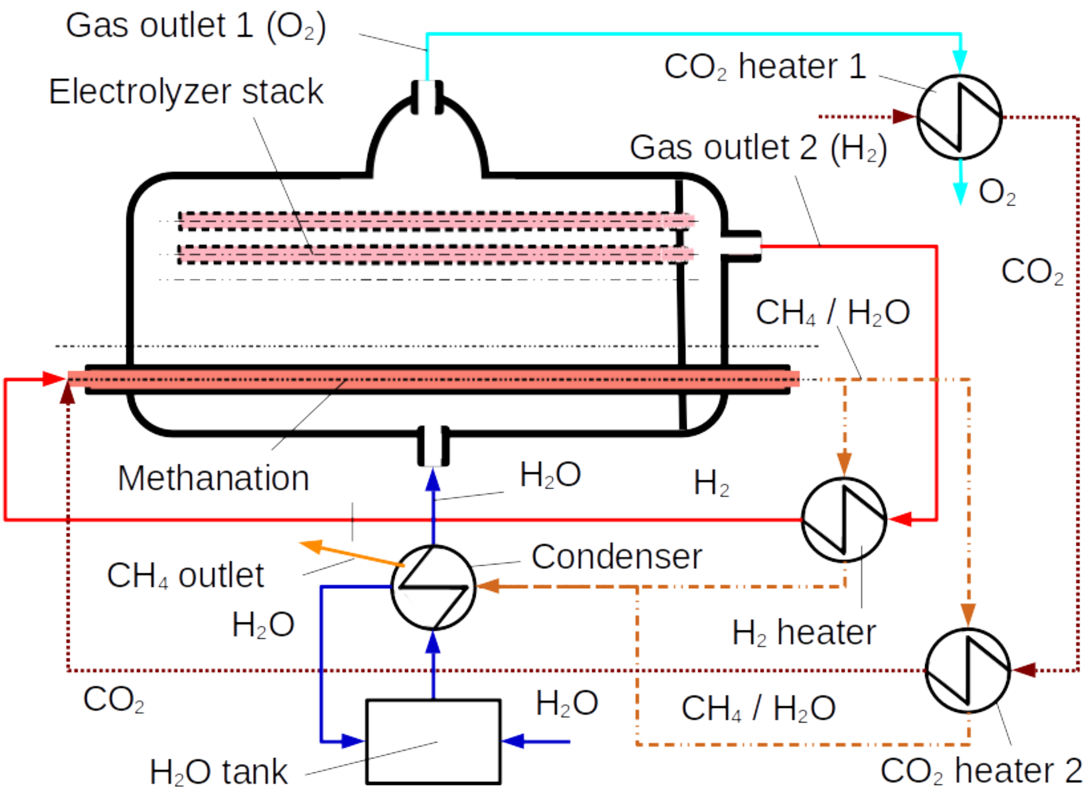


Figure 15. Principal BoP of Methanation with CO_2 as Reactant.

The principal structure of the heat recovery system is a consequence of the heat balance of the system and the Figure shows the principal heat recovery options of the system. The final design needs a detailed calculation for optimization also depending on the optimal methanation reactor temperature as e.g. depending on the catalyst and economic trade-offs. The temperature differences between the gas flows should be as small as possible from a thermodynamic point of view for the best approach of the reversible reference system. The technical realization needs higher temperature differences to avoid too large heat exchangers on the other side.

The incoming CO_2 can be heated up by the released O_2 in a first stage and by the CH_4/H_2O mixture leaving the methanation reactor in a second stage. The CH_4/H_2O mixture can by additionally used to heat up the incoming H_2. The condenser for the separation of CH_4 and H_2O is used for preheating the feed water. The possible process optimization as shown in **Figure 12** and its integration in the entire system depends also on a detailed engineering and is not considered here.

Using CO instead of CO_2 for the methanation following (2) in **Table 2** demands the integration of an additional CO_2 electrolyzer in an extra section of the steam generator. It also needs a different approach of heat recovery. More possibilities of system design can be found in the own work [15].

6. System Integration in Circular Economy

The targeted carbon cycle economy needs not only operating gas generators but also realized an entire CCC (closed carbon cycle). Thus the complete closed process chain needs to be discussed especially the carbon cycle including the CO_2 handling in the system. The experience from demonstration projects with the decarbonization of coal fired power plants was based on the Oxyfuel process where the coal was burnt with oxygen and flue gas recycling was used for the necessary cooling of the furnace. The high cost of the O_2 generation was a fundamental problem of the project cost. The proposed gas infrastructure merely represents a modification of existing plants, i.e. corresponds to TRL 8/9, fuel cell plants and Oxyfuel burners are still in an optimization phase in terms of costs and operating experience and are therefore classified as TRL 7/8.

The electrolysis as a main technology of the gas generation of CCC solves this cost problem because O_2 is a by-product and thus available for its utilization in combustion processes. Consequently, the future operation of combustion processes in a future carbon cycle economy is possible and can deliver CO_2 as raw material for CH_4 or C_2H_4 production. Fuel cells are the other CO_2 source in the cycle because the fuel utilization is combined with a separation of air and CO_2. The operation of the cycle as a component of the electric grid is explained in **Figure 16**.

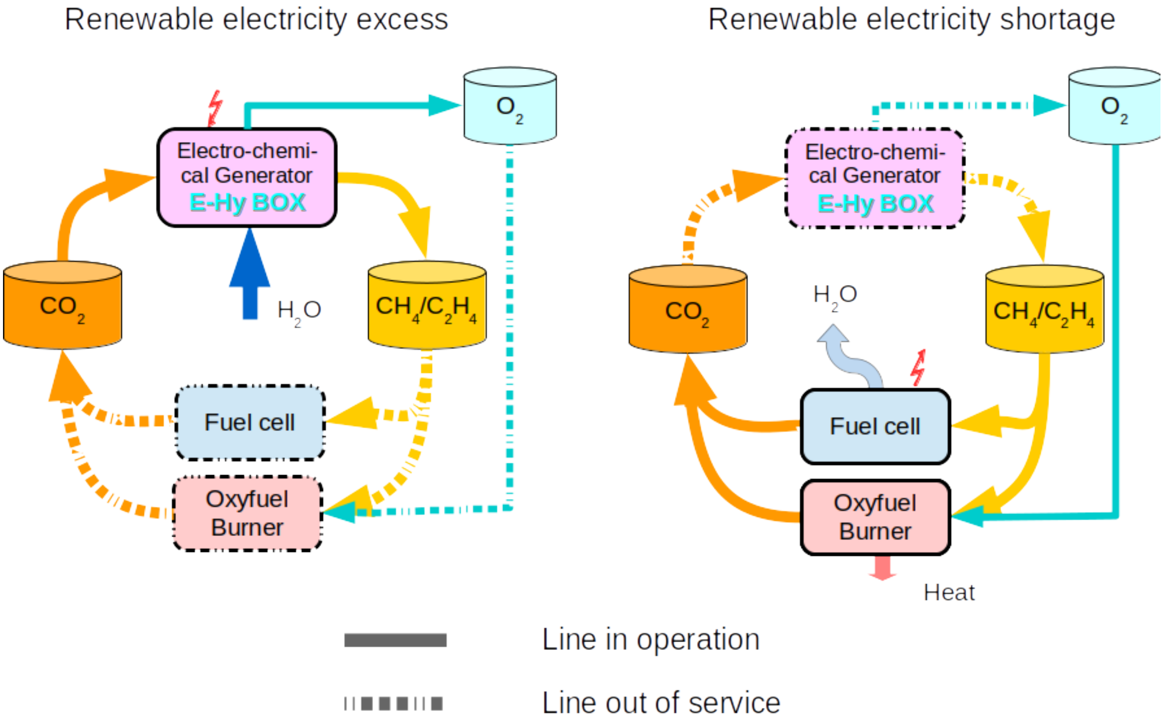


Figure 16. The Operation Modes of Closed Carbon Cycles.

The principal operation of CCC in the electric grid is defined by excess and shortage of renewable electricity. The excess operation is characterized by the electricity utilization of the electrochemical generators delivering CH_4, and C_2H_4 and being supplied with CO_2 from a storage facility and with H_2O from available sources. Possibly there might be the need in future to include H_2O in the cycle economy as well. The shortage operation is characterized by a shut down of the electrochemical generators and an operation of the installed fuel cells and Oxyfuel burners by utilizing the stored CH_4, and C_2H_4 for the production of electric power on demand of the electric grid.

However, CCC is the core of the carbon cycle economy but it has to be explained how the national economy and its facilities including industry and community can be integrated in the carbon cycle economy. This interacting combination of power supply, community, supply of industry with energy and raw material, generating H_2 as fuel for transportation or as raw material, and "green" hydrocarbons for combustion engines is presented in **Figure 17**.

The operation of CCC in power supply is only one part however an important one in the carbon cycle economy. Anyway it is the core of the entire system because renewable electricity is the only political targeted future source of energy however an increasing number of states see nuclear energy as a sort of renewable energy. The electrochemical CH_4, or C_2H_4 gas generators including the gas storage facilities for CH_4 or C_2H_4, and CO_2 are the second core component of the carbon cycle economy. Gas generators and electric grid are connected to all energy consumers by supply lines. The electricity supply follows the demand as described in **Figure 1** by using the storage capacity of CH_4 or C_2H_4. The supply lines are needed for the transport of electricity, the fuels H_2, CH_4, and C_2H_4, and the raw materials CO_2, H_2O, O_2 needed or produced by the gas generators. The industrial production can be supplied with all these substances as raw material for its production. The system can supply the transportation sector with hydrogen and also "green" fuel. The carbon circular economy can also merge with hydrogen economy as today the natural gas is used for generating H_2 by reforming the natural gas. The produced CO_2 is recycled within the CCC process. Another option is the use of 'green CO_2' emitted in the flue gas of biogas plants. A delivered quantity of 'green CO_2' can be used to produce "green" CH_4 or C_2H_4 for direct use in combustion engines or for the production of green fuel.

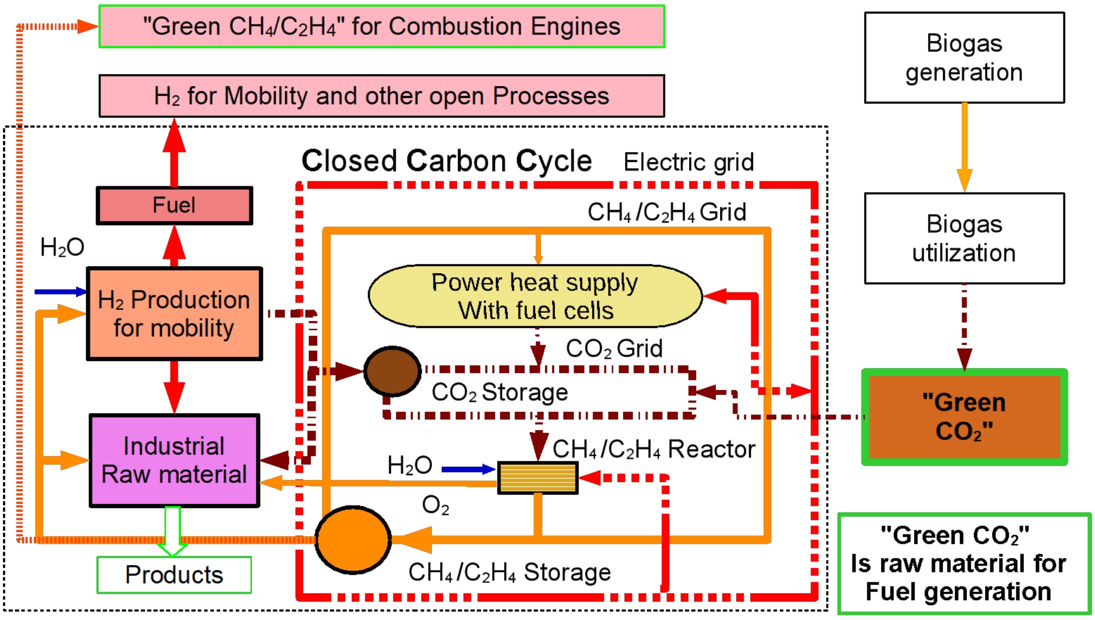


Figure 17. Interaction of Community, Industry and Mobility in a Future Carbon Cycle Economy.

7. Economy and Performance of Integrated System

The economy of any energy system can be expressed by the relation of fixed and variable cost and the delivered quantity of energy. The share of fixed costs is the quotient of the capacity-related acquisition value (specific investment) and the product of annual full-load operating hours and depreciation period.

The associated share of variable costs is the quotient of the electricity price and the conversion efficiency of the gas generator. The conversion efficiency of the gas generator is the ratio of the generated thermodynamic potential of the gas and the electrical work input. These considerations are important for an optimal system design but they don't include the influence of financing, because important aspects as the price of CO_2 and the byproduct O_2 are still open as discussed in the own analysis [8].

As shown in there, the specific costs can be summarized in such a way that the two main shares of the fixed and variable costs mentioned can be divided into a technology-determined share and a correction share. The correction shares are company-specific empirical values in the order of about 5 %, to which the influence of capital costs needs to be added. This makes it possible to estimate the trends in the cost relevance of the above-mentioned parameters in an initial approach, if essential design and financing data are still open. **Figure 18** summarizes these results into an overall view. This consideration is not directly dependent on the gas quality but only via its influence on the process parameters.

The fixed cost share of the gas production costs depends linearly on the capacity-related acquisition value (here 500 €/kW). It decreases significantly with increasing depreciation period and increasing annual full load operating hours. A value of less than 1 ct/kWh is reached here between 2000 and 5000 full load hours, depending on the depreciation period. The electricity price and the conversion efficiency are the main influences on the variable cost component of the gas production costs. The minimum electricity price was set at 2 ct/kWh, forming its lower limit, which is more than twice as high as realistic values for the fixed cost share.

High annual full load hours at a low electricity price and a high conversion efficiency are the leading variables for favorable gas production costs. In contrast, deviations in the purchase value are of somewhat less relevance especially at high full load hours and long depreciation times. It can be further optimized in a learning curve. The necessary minimum combination for the production of a low-cost syngas consists of the combination of low-cost electricity and high full-load hours of the state of the art gas generators. The plot of full-load operating hours in Figure18 shows that currently only nuclear energy is available as an economic option because only it can combine high full-load hours with low-cost electricity. Of the renewable energies, only solar energy has a prospect of being able to provide similarly favorable electricity prices in the future but the annual full load hours are low [16]. A possibility to increase the full load hours could be a solar belt consisting of connected solar cells crossing meridians. The use of bioenergy, which is also theoretically conceivable, is not expedient because it would limit the portfolio of available renewable energy instead of expanding it. The use of batteries for grid stabilization is not only limited by their capacity [17], but is also uncompetitive in terms of cost compared to adapting the existing natural gas infrastructure.

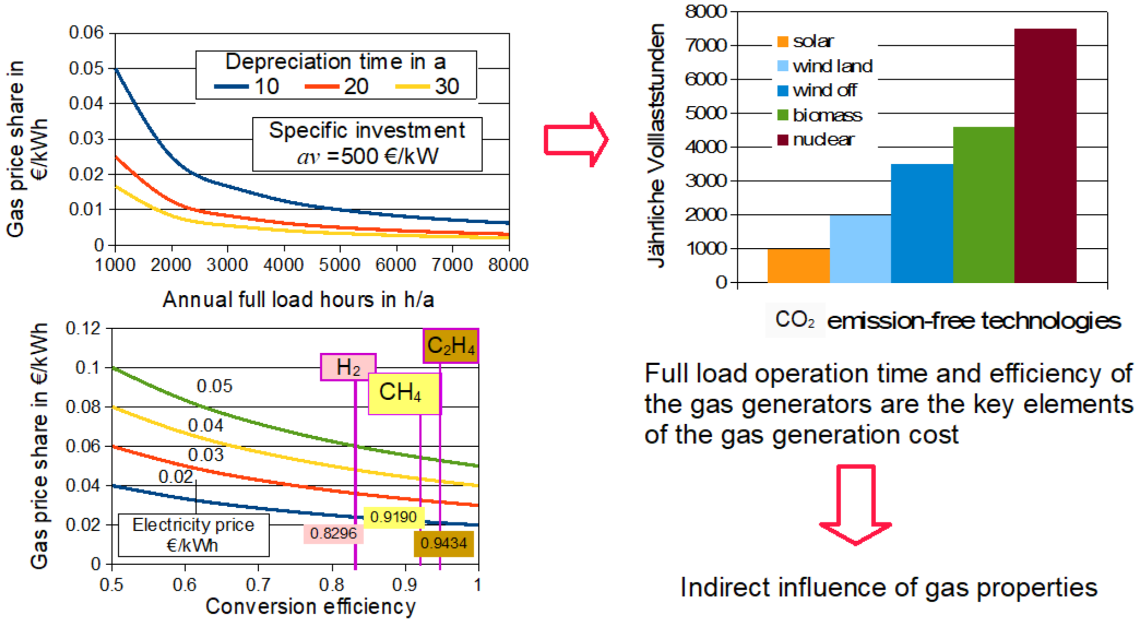


Figure 18. Economic Feasibility and Operation and Design Parameters [8].

The grid stabilizing power generation based on synthetic gases needs a high efficiency, a high flexibility and a high availability as well. **Figure 19** explains the possible solutions. There, above the charging efficiency, i.e. the efficiency of gas generation, the discharging efficiency, i.e. the efficiency of power generation, is entered. The total efficiency of a cycle is then the product of these two partial efficiencies. Because of the problems shown with thermal processes for the generation of CH_4 or C_2H_4, the efficiencies of their generation are similar to those of hydrogen electrolysis at around 70% or even lower [14], although theoretically the molecules of CH_4 or C_2H_4 have an evaporation loss of between about 6 and 10% because of their lower content of H_2. However, with the help of the solutions shown above, efficiencies of about 90% can be expected for improved gas generation, and efficiencies of about 80% have been calculated for electricity generation based on SOFC-GT (Solid Oxide Ful Cell– Gas Turbine combination), which is a highly efficient development presented in the own work [18]. While the efficiencies of gas turbine combined cycle power plants are currently around 65% following KND (Klimaneutrales Deutschland) [19]. Thus, with the help of this new combination of gas generation and electricity generation, an efficiency can be expected which, at about 70%, corresponds to that of pumped storage power plants, which is about a 20% increase in efficiency above that of today's PtG technology with less than 50% for regenerative power generation.

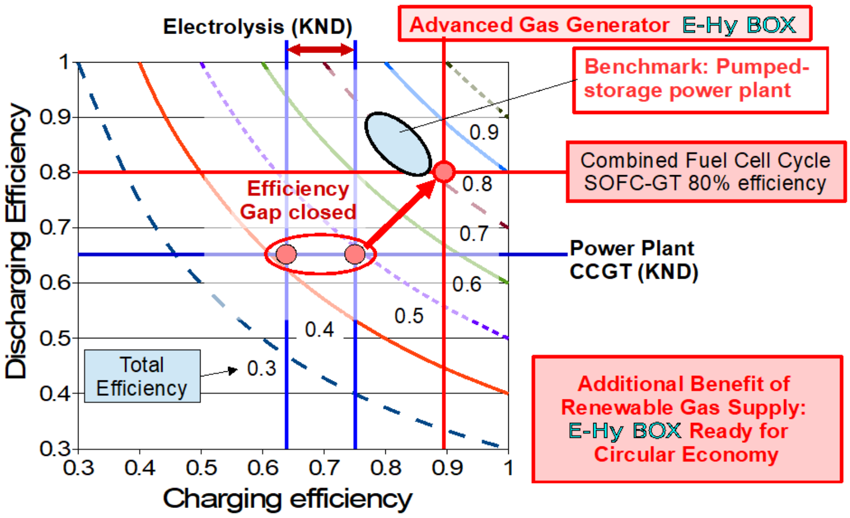


Figure 19. The Efficiency of Power Generation from Synthetic Gas Compared with Pump Storage Hydro Power Plants.

8. Conclusions

As already mentioned, the carbon cycle economy using CCC requires closed cycles in order not to emit CO_2 and to keep it as reactant in the loop. However, this is not suiTable for open systems that release waste gas into the atmosphere. Consequently, H_2 or another "green" fuel must always be used. As shown above, both hydrogen storage and also long-distance transport are expensive because of its low volumetric energy density. This means that with H_2 only less than a quarter of the energy currently stored with natural gas (CH_4) is available and therefore no secure supply can be expected even if the KND scenario is implemented [19]. The solution is to use C_2H_4, which can store more than four times as much energy as CH_4 in the same volume. The demand for a secure energy supply therefore requires a carbon cycle economy. A viable overall concept must, on the one hand, ensure security of supply with large energy reserves and favorable storage and transport costs, and, on the other hand, also meet the demand for hydrogen where it is needed. **Figure 20** shows the schematic of a suitable overall concept.

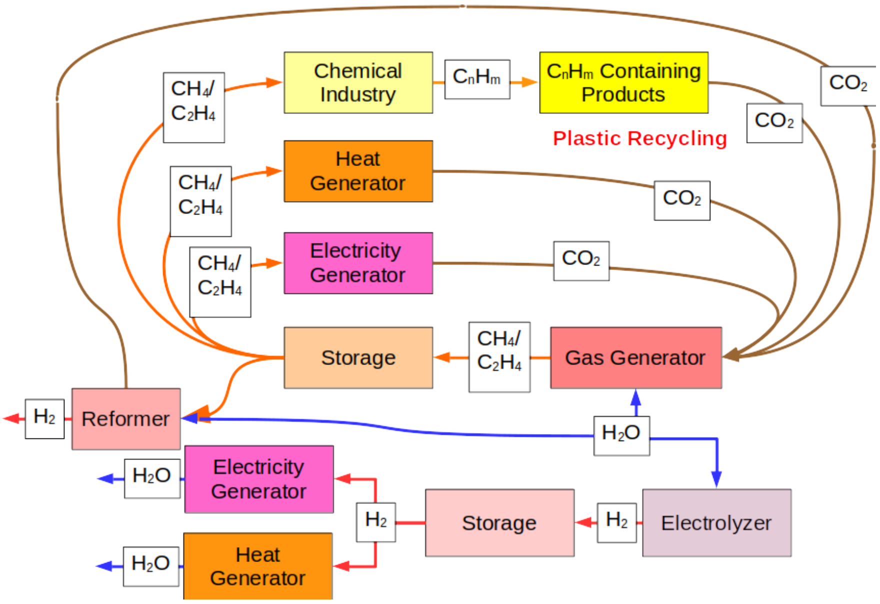


Figure 20. Vision of a Future Carbon Cycle Economy.

The use of the CCC system can significantly reduce the transport and storage costs for all the synthetic gases required and significantly increase energy security. The industrial plants can continue to operate with largely compatible gases. Although conversions and adaptations may be required, they are likely to remain within limits. This classification is a consequence of the studies presented here, which clearly show that H_2 would not satisfactorily meet any single requirement with respect to storage and transport and would incur considerable additional costs. Conversely, however, since H_2 is needed, a solution is required that exploits opportunities inherent in the system for optimizing operations and reducing costs. Today, H_2 is produced from natural gas on an industrial scale, and this process can be adopted here. This allows to always use the advantages of hydrocarbons for energy transportation and storage and then to extract H_2 via reforming processes as today. This does not prevent local H_2, production by electrolysis if economical, but expands the technical options for end users. In addition to the known transport applications, it is, for example, a question of economic viability whether rural individual houses should then be heated with H_2, CH_4 or C_2H_4 and then CHP (Combined Heat and Power) may make sense. Which applications prove to be economical and when is not the subject here. The aim here is to enable users to continue to have a secure and affordable energy supply without major additional costs, but also to enable an openness of technology for end customers without additional costs in the question of secure energy reserves and cost-effective energy transport. Although CCC offers clear advantages over a hydrogen economy, the costs must be measured against the global energy market. The results in the work of Winkler indicate that with favorable but achievable electricity prices and good gas production efficiencies [8], competitiveness with liquefied natural gas can be achieved. If gas storage facilities are used for reconversion to electricity, it should be possible to achieve efficiency values comparable to pumped storage power plants. Nevertheless, further detailed studies are required to optimize the economic viability. These requirements necessitate the implementation of the analyses presented here in marketable products in the field of solar power generation, gas generation and reconversion to electricity. The development of photovoltaics is promising, as shown by falling market prices, but is not the focus of this article. For the industrial production sector, an increase in production efficiency to further reduce costs is certainly an important driver here. In gas generation, a distinction must be made between two areas: system design (BoP) (TRL 7–9) and integrated reaction technology (TRL 2–4). In order to minimize the risk of the system design, proven components of classic steam technology were used to a large extent, in order to limit the entire development risk to the reaction technology as far as possible. The following main topics can be mentioned here: adaptation of the electrolysis to changed operational management; for the second reaction stage, these are the catalyst development, the integrated oxygen ion-conducting electrolysis, the fuel cell injector and the integrated heat recovery. The implementation of the existing concepts of SOFC-GT power plants (TRL 4–5) in test plants and their further experimental development is of great importance for a high degree of efficiency in reconversion to electricity. Basic research can certainly make important contributions in the field of quantum technology for both photovoltaics and electrochemical components, but should also consider the subsequent implementation in industrial processes, such as the selection of materials and their supply chains, from the outset. The reports of the German Federal Audit Office and the European Court of Auditors clearly show that the German energy transition and the EU's "Green Deal" have reached their limits and need to be restarted [20–22]. The carbon cycle economy can make a significant contribution to reducing costs because improving energy efficiency cannot contribute to reducing CO_2 emissions, as the cycle itself does not emit CO_2. This means that the investment costs of the carbon cycle economy alone are sufficient to achieve climate protection targets. According to the own work [23], the investment costs of the carbon cycle technology will be less than €2 trillion based on the cost basis according to the Agora edited study by Prognos et al. [19], while the current approach according to the Agora edited study by Prognos et al. [19], including extensive energy efficiency improvements, could cost as much as €10 trillion. Improvements in energy efficiency can then result according to market economy principles and not legal requirements.

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Institutional Review Board Statement

Not applicable.

Informed Consent Statement

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Data Availability Statement

Data supporting reported results can be found in reference [15].

Conflicts of Interest

The author declares no conflict of interests. Patent pending.

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