

Energy and greenhouse balance of photocatalytic CO₂ conversion to methanol

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Abstract. Within the Leading-Edge Cluster “Forum Organic Electronic”, the research project “Solar2Fuel” funded by the German Ministry of education and research (BMBF) (2009 – 2012), EnBW, BASF, Karlsruhe Institute of Technology and Ruprecht-Karls-University of Heidelberg aim to develop a future solar powered CO₂ to methanol conversion technology. CO₂ from stationary sources such as power plants shall be catalytically converted together with water to a product such as methanol by use of solar irradiation. For this purpose a catalyst shall be developed. EnBW investigates the required boundary conditions to make such a principle interesting with respect to energy and greenhouse gas balance as well as economic evaluations. The assessment of boundary conditions includes the analysis of the whole chain from power generation, CO₂ capture and transport, a virtual photocatalytic reactor, the product purification and use in the traffic sector. Most important technical factors of the process such as CO₂ conversion efficiency is presented. CO₂ capturing and liquefaction are the most energy intensive process steps, CO₂ transport in pipeline is highly energy efficient and depending on energy need of the photoconversion step and the product purification, the overall greenhouse gas balance is comparable with the underground storage of the captured CO₂.

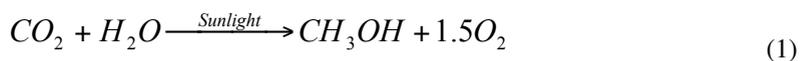
1 Introduction

Principle of photocatalytic CO₂ conversion into methanol

The research project “Solar2Fuel” pursues the future vision of photocatalytic CO₂ conversion, to use direct sunlight as energy resource for reduction of CO₂. Thus, CO₂ can be used as feedstock for renewable fuel and energy storage. CO₂ captured from flue gas of a power station is liquefied and transported to the location of a photocatalytic reactor field, which converts CO₂ with sunlight to methanol. The latter can be used as fuel in cars and therefore substitute oil based gasoline and subsequently avoid emissions from fossil gasoline.

Photocatalytic reduction of CO₂ is a redox-reaction with water as reducing agent. The net chemical reaction is shown in equation (1).

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EnBW investigates the required boundary conditions for an environmentally friendly and economic operation of the “Solar2Fuel” process, if captured CO₂ is transported far from Germany to sunny regions in South Africa (Figure 1).

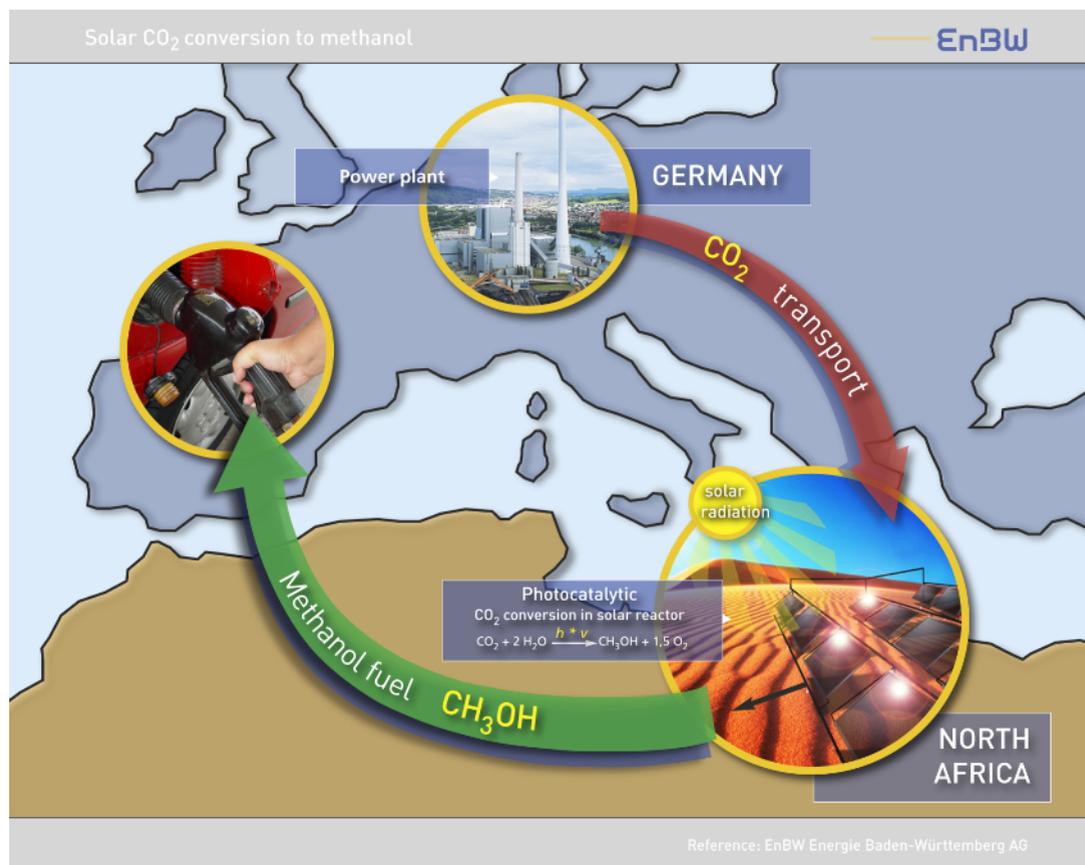


Figure 1: The “Solar2Fuel” system. Captured CO₂ in Europe, transport to sunny regions in South Africa for photocatalytic conversion

The assessment of boundary conditions includes the analysis of the whole chain from power generation, CO₂ capture and transport, a virtual photocatalytic reactor the product purification and use in the traffic sector. It is evaluated, if it is energetically and economically feasible to transport huge amount of CO₂ from Germany to sunny areas in South Africa.

2 The process chain and data of photocatalytic CO₂ conversion into methanol

In the following, data for CO₂ supply to the reactor field, beginning with CO₂ capture and liquefaction in fossil fuelled power plants in Germany, subsequent pipeline transportation to the place of delivery in In Salah (South Africa) are presented.

The cost effectiveness and the efficiency of CO₂ pipeline transport at far distance increases strongly with larger inner diameter of the pipeline. Half inner diameter gives rise to 16 fold increase in

friction loss (Hagen Poiseuille theory). Thus, the pipeline is simulated with a large inner diameter of 1.1 meter and 50 million tons annual capacity. This annual amount of captured CO₂ corresponds to 6.397 GW_{el} coal fired power stations with a net efficiency of 34.3 % after CO₂ capture and liquefaction and CO₂ transport.

We assume that 90% of the delivered CO₂ is converted to methanol, 10% is lost to the atmosphere during the conversion process. Oil based gasoline can be substituted by methanol on basis of lower heating value. The higher efficiency of methanol combustion compared to gasoline combustion is not considered in this study.

2.1 Carbon capture and liquefaction in fossil fuelled power stations

As explained above, the annual transportation capacity of the CO₂ pipeline is set to 50 million tons per year. The total thermal power of power stations with carbon capture is set to 18.653 GW_{th}, to operate the pipeline with full capacity, 8760 hours per year. That gives rise to a total net power of 6.397 GW_{el} of power stations after losses due to carbon capture, liquefaction and transport.

To compare the capture case (“Solar2Fuel system) with the today’s situation of power stations without carbon capture and oil based gasoline fuelled cars, the capacity of the power stations without carbon capture is set to 6.397 GW_{el} as well (Table 1).

Table 1: Data about power stations and CO₂ capture & liquefaction

Category	Unit	Power station without carbon capture	Power station with carbon capture
thermal power	[GW _{th}]	14.027	18.653
Net efficiency without carbon capture	[%]	45.6	45.6
Efficiency drop due to carbon capture with MEA ¹	[%]	-	-7,4
Efficiency drop due to liquefaction	[%]	-	-3,1
Efficiency drop due to pipeline transport	[%]	-	-0.76
Net Efficiency after carbon capture	[%]	-	34.3
Electricity output	[GW _{el}]	6.397	6.397
Assumed annual operating hours	[h]	8,760	8,760
CO ₂ capture efficiency	[%]	-	90
Annual amount of captured CO ₂	[million t]	-	50.0
Annual CO ₂ emission at power station	[million t]	41,779	5.556
Energy demand for CO ₂ liquefaction ¹	[kWh _{el} /t]	-	102.3

¹=calculated with formula after [01], compression to 8 MPa, 4 compression stages with intercooling

2.2 CO₂ pipeline transport from Germany to North Africa

Data for a CO₂ pipeline from Germany to Algeria via Italy, Sicily and Tunisia is listed in **Table 2**. Analysis and evaluation was done from RBS wave GmbH and are taken from [02, 03]

Table 2: Data about CO₂ pipeline from Germany to Algeria

Category	Unit	Number
Length of the pipeline	[km]	3,743
Inner diameter of the pipeline	[m]	1.1
Required amount of steel	[t]	3,199,496
Total pressure loss due to friction	[bar]	662
Electricity consumption for pumping stations per t CO ₂	[kWh _{el} /t]	24.74
Annual CO ₂ transport capacity	[million t]	50.0

2.3 Comparison of energy and greenhouse gas balance of today's situation and the "Solar2Fuel" scenario

2.3.1 Required data for evaluation of the system balances

For sound comparison of today's situation and the "Solar2Fuel" system, both cases need to supply the same amount of electricity to the grid as well as the same amount of fuel (measured in lower heating value) to combustion engines in the traffic sector. Data for lower and higher heating value for gasoline and methanol is listed in **Table 3**.

Table 3: Higher and lower heating value from gasoline and methanol

Category	Unit	Gasoline	Methanol	Reference
Density	[kg]	0.726	0.79	[04, 05]
Lower heating value (LHV)	[kWh/kg]	12.08	5.53	[04, 05]
Higher heating value (HHV)	[kWh/kg]	12.92	6.30	[04]
CO ₂ emission after combustion	[kg/kg]	3.178	1.375	-

1.375 kg CO₂ yield one kg methanol after photocatalytic conversion. With an assumed loss of 10% CO₂ within the conversion process, 1.528 kg CO₂/kg methanol need to be delivered to the photocatalytic reactor field and 32.727 million tons methanol are produced (**Table 4**). Based on lower heating value, the methanol can substitute 14.994 million tons gasoline.

Table 4: Data about photocatalytic CO₂ conversion

Category	Unit	Amount
Annual CO ₂ supply to the place of CO ₂ conversion	[million t]	50.000
Stoichiometric CO ₂ consumption for methanol production	[kg/kg]	1.375
Specific CO ₂ consumption/kg methanol, 10% CO ₂ loss considered	[kg/kg]	1.528
Annual methanol production	[million t]	32.727
Annual substituted amount of gasoline	[million t]	14.994

To analyse the greenhouse gas balance of both systems, the upstream emissions of coal mining and supply, gasoline supply and the construction of the power stations as well as of the CO₂ pipeline are taken into account. Specific values for the commodities, energy carrier and the infrastructure (power station and pipeline) are listed in **Table 5**.

Table 5: Greenhouse gas emissions (GHG) and cumulated energy demand (CED) due to upstream chains

Category	Unit	Number	Reference
CED of coal supply	kWh/kWh	0.09	[06, 07]
GHG emission of coal supply	g CO ₂ -eq ¹ /kWh	46.4	[06, 07]
CED of gasoline supply	kWh/kWh	0.202	[08]
GHG emission of gasoline supply	g CO ₂ -eq/kWh	56.5	[08]
CED of steel for pipeline and its construction ²	kWh/kg	7.0	-
GHG of steel for pipeline and its construction ³	kg CO ₂ -eq/kg	2.7	[09]
CED of power station construction ⁴	Wh/kWh _{el} (without/with carbon capture)	5.7/7.5	-
GHG of power station construction ⁵	g CO ₂ -eq/kWh _{el} (without/with carbon capture)	2.2/2.9	[10] according to Hondo
CED for CO ₂ capture facility ⁶	Wh/kWh _{el}	7.5	-
GHG for CO ₂ capture facility ⁶	g CO ₂ -eq/kWh _{el}	2.9	-

¹= "CO₂-eq", add on "-eq" denotes CO₂-equivalent

²= estimation by dividing GHG with specific emission factor of hard coal (2.7/0.3864).

46.4 g CO₂-eq/kWh origins from coal supply chain, 340 g/kWh from combustion

³= data from Probas for steel; 2.0 kg CO₂-eq/kg steel plus 15% for manufacturing of tube segments and another 20% for pipeline construction itself

⁴=estimation by dividing GHG for construction with 386.4 g CO₂eq/kWh primary energy (340 g CO₂/kWh LHV + 46.4 g CO₂-eq from coal supply chain)

⁵=data taken from [10] according to Hondo, adapted to capacity utilization

⁶=estimated in the same order of magnitude as the power station itself

2.3.2 Results about annual greenhouse gas emissions

Table 6 shows the greenhouse gas emissions for the today's situation and the "Solar2Fuel" system including the contribution of the different steps of the process chains.

Table 6: Annual GHG emissions of today's situation and the "Solar2Fuel" system

Category	Unit	Today's Situation	"Solar2Fuel" System
Annual GHG emission in upstream chain of coal supply (mining and transportation)	[million t CO ₂ eq]	5.707	7.588
Annual GHG emission from power station construction (incl. CO ₂ capture)	[million t CO ₂ eq]	0.123	0.328
Annual GHG emission from direct CO ₂ emission in power station	[million t CO ₂ eq]	41.779	5.556
Annual GHG emission from pipeline construction ¹	[million t CO ₂ eq]	-	0.254
Annual GHG emission via CO ₂ loss in photocatalytic CO ₂ conversion process to methanol	[million t CO ₂ eq]		5.000
Annual GHG emission in upstream chain of gasoline supply (drilling, refining, transportation)	[million t CO ₂ eq]	10.233	-
Annual GHG emission via gasoline combustion	[million t CO ₂ eq]	47.650	-
Annual GHG emission via methanol combustion	[million t CO ₂ eq]	-	45.000
Total annual GHG emission of the system²	[million t CO₂eq]	105.492	63.726
Relative reduction of the "Solar2Fuel" system	[%]	-	39.6%

¹= 35 years pipeline lifetime assumed

²=upstream chains of construction of photocatalytic reactor is not considered as well as the methanol purification process. Final relative GHG emission reduction will be lower.

The relative annual emission reduction of the "Solar2Fuel" system compared to the today's situation is about 39.6%. It is to notice, that the emissions for the construction of a reactor field for the photocatalytic reaction is not included in the calculation as well as the purification of the methanol to fuel grade.

To estimate the effect of methanol purification to fuel grade, we assume that the heat input for distillation is about 25% of the lower heating value of methanol and that the required heat is supplied with steam supplied via parabolic trough collector.

Table 7 shows the CED and GHG for solar thermal electricity supply from solar concentrating power plants with parabolic trough [11, 12].

Table 7: CED and GHG of electricity supplied from solar thermal power station with parabolic trough

Category	Unit	Number
CED for supply of one kWh _{el} by solar thermal power station	[MJ/kWh _{el}]	0.14
GHG emissions for supply of one kWh _{el} by solar thermal power station	[g CO ₂ -eq/kWh _{el}]	14

Supposed that the efficiency of steam/heat conversion into electricity is 30%, it can be concluded, that the CED for one kWh steam is only $0.14 \cdot 0.3 = 0.042$ MJ or 11.6 Wh/kWh.

Considering GHG emission, that means 4.2 g CO₂-eq/kWh steam.

This gives rise to GHG emissions of 23.2 kg CO₂-eq/t purified methanol or 760,124 t CO₂-eq every year. The "Solar2Fuel" system has a relative GHG reduction of 38.84% compared with today's situation. It can be concluded, that purification will have only little effect, as long as the required heat is supplied with solar energy.

2.3.3 Results about annual primary energy consumption

Table 8 shows the annual primary energy consumption of today's situation and the "Solar2Fuel" system.

Table 8: Annual primary energy consumption of today's situation and the "Solar2Fuel" system

Category	Unit	Today's Situation	"Solar2Fuel" system
Annual CED in upstream chain of coal supply (mining and transportation)	[TWh]	11.059	14.706
Annual CED from power station construction (incl. capture facility)	[TWh]	0.319	0.849
Annual primary energy consumption through coal consumption in power station	[TWh]	122.880	163.399
Annual CED from pipeline construction ¹	[TWh]	-	0.639
Annual CED in upstream chain of gasoline supply (drilling, refining, transportation)	[TWh]	36.587	-
Annual primary energy consumption via gasoline combustion	[TWh]	181.122	-
Total annual primary energy consumption of the system²	[TWh]	351.968	179.593
Relative reduction of the "Solar2Fuel" system	[%]	-	49.0%

Table 8 does not include the CED of the upstream chain for construction of parabolic trough field for steam generation of methanol distillation. The supply of 45.29 TWh steam for distillation does roughly cause a CED for construction of annual 0.525 TWh and would decrease the relative advantage of “Solar2Fuel” system to 48.8%.

Here is to mention again, that CED for construction of reactor field for photocatalytic conversion process is not included as well as the energy for operation of such a field.

2.4 Comparison of the relative emission reduction of “Solar2Fuel” system with carbon capture and storage (CCS)

Under environmental aspects, the “Solar2Fuel” system does only make sense, if there is an advantage compared to carbon capture and storage of CO₂ in underground formations.

In [13] is described, that the greenhouse gas emissions for the production of one kWh_{el} including the upstream processes can be reduced by 71.4%. In this case, cars will further use oil based gasoline, while in the “Solar2Fuel” system, the captured CO₂ is not stored in underground formations, but converted back to methanol and substitutes gasoline in cars. With data from **Table 6** it can be concluded, that GHG emission reduction in the system with CCS and oil based gasoline consumption for cars, is about -32.2% compared to the today’s situation.

The comparison of the relative greenhouse gas emission of the „Solar2Fuel“ system with carbon capture and storage is shown in **Figure 2**. On axis of abscissa, the CO₂ conversion rate is depicted. It describes the percentage of CO₂ supplied to the reactor field, which is converted to methanol. The rest is lost to the atmosphere. The graph shows, that greenhouse gas balance of “Solar2Fuel” system can be better than CCS, if CO₂ conversion rate of supplied CO₂ to the reactor field is above 77%. The blue line describes the relative GHG reduction of “Solar2Fuel” system compared to the today’s situation. The red line is independent of CO₂ conversion rate and shows the relative GHG reduction of CCS case compared to the today’s situation. The blue line of “Solar2Fuel” system in **Figure 2** does again not include the upstream chain of photocatalytic reactor field construction and methanol purification. Thus, blue line will shift down/to the right side, if these two factors are considered. To compete with the CCS case, the minimum CO₂ conversion rate needs to be higher than 77%.

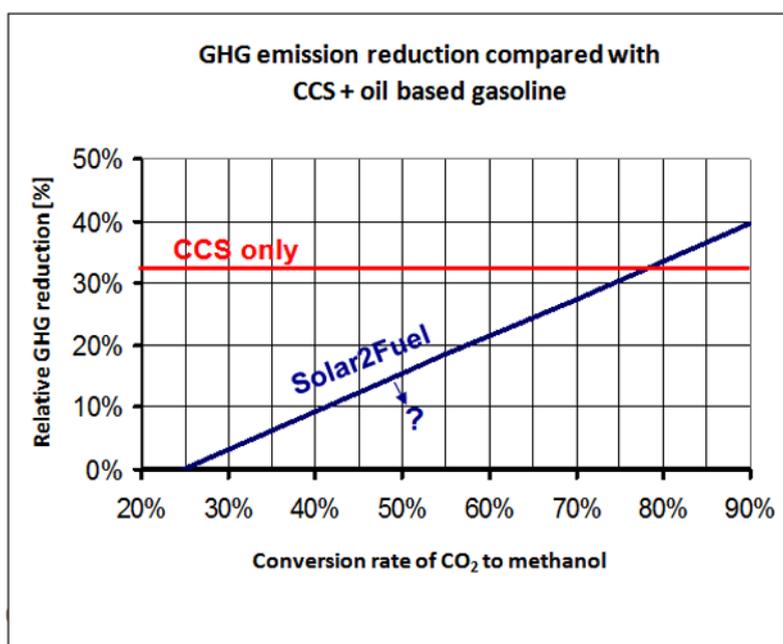


Figure 2: Relative GHG emission reduction of "Solar2Fuel" system and CCS case (CCS and oil based gasoline) compared to today's situation

3 Economic aspects of the "Solar2Fuel" system

The economic feasibility of solar CO₂ conversion to methanol depends of a variety of parameters. **Table 9** lists some important parameters that do influence the profitability of photocatalytic methanol production.

Table 9: Parameters that influence the profitability of solar CO₂ conversion to methanol

Category	Unit	Description
Solar irradiation	[kWh/m ² *a]	The more annual solar irradiation, the higher is the area specific methanol yield and thus generates higher cash flows. In all subsequent calculations, a solar irradiation of 2,000 kWh/m ² /year is assumed.
Photo conversion efficiency (PCE)	[%]	Relative energy of solar irradiation, that is stored as chemical energy in methanol (based on HHV)
CO ₂ supply cost	[€/t]	The cost of CO ₂ supply to the photocatalytic reactor field will strongly influence methanol production cost
Carbon dioxide conversion rate	[%]	Part of delivered amount of CO ₂ , that is converted into methanol
Energy demand for operation of photocatalytic reactor field	[TWh]	Energy consumption for reactor field operation will influence energy and greenhouse balance. Furthermore energy has a price and has to be

		financed by sale of the product (methanol)
Capital expense for reactor field construction	[€]	Depending on the PCE, solar CO ₂ conversion needs huge area. The area specific invest together with interest rates will have strong influence on production cost
Heat requirement for methanol purification	[kWh/t]	It is expected, that methanol leaves the reactor in low concentrated in a mixture with water. The higher the heat requirement, the higher the production cost of solar methanol

Table 10 shows the underlying assumptions in the economic evaluation of photocatalytic methanol production from CO₂.

Table 10: Parameters for economic evaluation of photocatalytic methanol production from CO₂

Category	Unit	Number	Reference
CO ₂ capture & liquefaction cost	[€/t]	32	[14]
CO ₂ transportation from Germany to North Africa via 3,743 km	[€/t]	13.7	[02]
Cost of CO ₂ over night storage in pipe storage system ¹	[€/t]	0.32	[02]
Total CO ₂ supply cost	[€/t]	46.0	[02]
Electricity consumption for photocatalytic reactor operation	[MWh/ha*a]	150	-
Electricity cost	[€ct/kWh _{el}]	5.0	-
Heat requirement for product distillation to fuel grade for one ton methanol (assumption) =25% LHV	[kWh/t]	1,383	-
Cost of solar heat supply	[€ct/kWh]	2.0	-
Depreciation time for the photocatalytic reactor field in years	[a]	20	
Interest rate for capital expenditure	[%]	10	
Solar irradiation	[kWh/m ² /a]	2,000	

¹= photocatalytic reactor field operates only during daytime. CO₂ arriving during the night period needs to be stored. Storage capacity is set to 14 hours pipeline capacity with pipe storage of 1.4 m diameter and specific invest of 3,000 €/m.

Figure 3 shows the methanol yield per hectare in dependence of photo conversion efficiency from 1% to 20%. This hectare specific yield is used for the following evaluation.

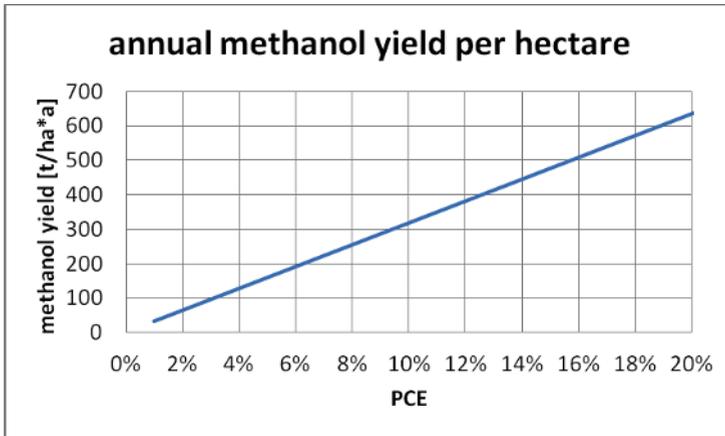


Figure 3: Methanol yield per hectare in dependence on photo conversion efficiency (PCE)

Figure 4 shows the hectare specific monetary turnover from sales of the product methanol for different price. From this money flow, the construction and operation of the photocatalytic reactor field need to be financed.

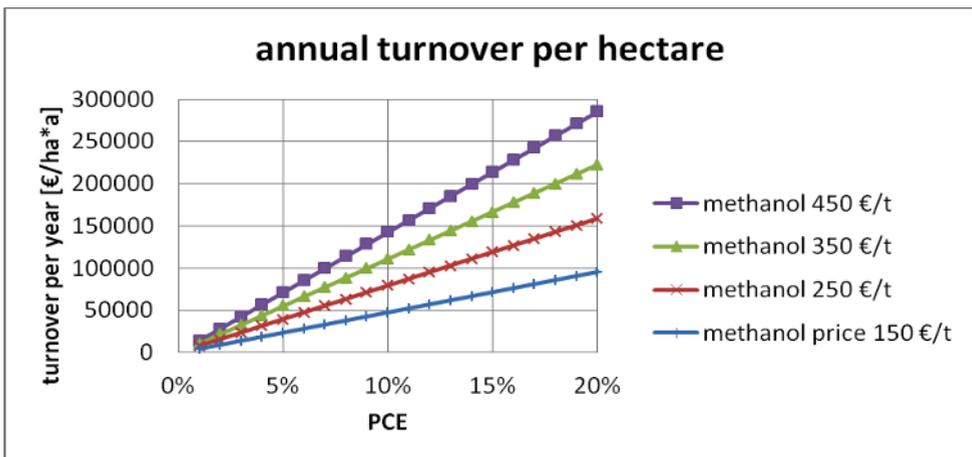


Figure 4: Hectare specific monetary turnover for different methanol prices

The construction has an important impact on the methanol production cost because the solar energy powered CO₂ conversion is a process with intensive land use. Subsequently, the hectare specific capital expenditure needs to be as low as possible. **Figure 5** shows the decrease of the free cash flow that can support the capital expenditure after considering the cost of CO₂ supply (46.0 €/t), electricity cost for reactor field operation (assumptions: 150 MWh/ha*year; 5 €/kWh_{el}) and cost for solar steam generation (1,383 kWh/t methanol for 2.0 €/kWh). These cost factors reduce the free cash flow about one third.

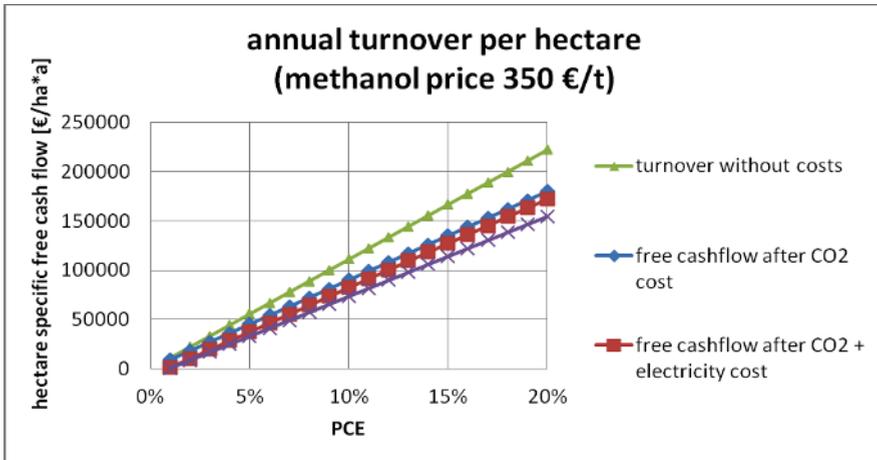


Figure 5: Effect of CO₂-, electricity- and solar heat cost on hectare specific free cash flow

The maximum allowed hectare specific invest, that can be financed with the free cash flow can be evaluated with the cash method. **Figure 6** shows the hectare specific net present value of photocatalytic reactor field. The blue horizontal line introduces a benchmark to get an imagination of the minimum required capital expenditure for the construction of a photocatalytic reactor field. A photovoltaic field with a land cover ratio of 70% and panel efficiency of 20% has a hectare specific peak capacity of 1,400 kW_p. With an assumed future photovoltaic system cost of cheap 500 €/kW, this gives rise to hectare specific capital expenditure of 700,000 €. It can be expected, that the construction of a reactor field for photocatalytic CO₂ conversion is rather more expensive than an easy and simple photovoltaic system. A photocatalytic reactor field need to have a transparent glass cover to prevent evaporation and CO₂ loss. Behind the transparent glass a catalyst absorbs the light. The field need to include a good network of pipes to handle liquid and gaseous flows together with sensors and pumps. Solar glass alone cost already 10 €/m² [15]. That is equivalent to 100,000 €/hectare aperture area photocatalytic reactor field and already 14% of the benchmark of 700,000 €/hectare.

Therefore, it can be considered as likely, that the hectare specific net present value of a photocatalytic reactor field need to exceed the investment for photovoltaic field on one hectare.

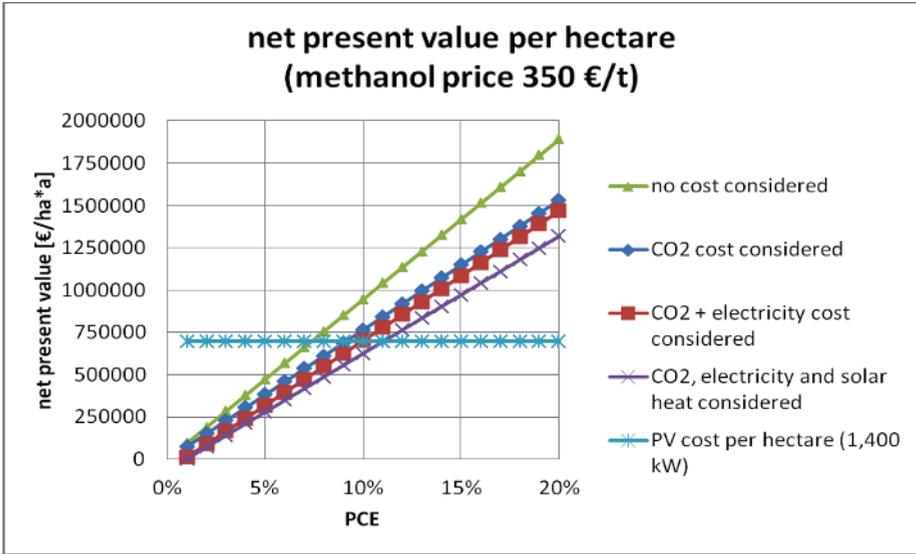


Figure 6: Hectare specific net present value calculated with methanol price of 350 €/t in dependence on PCE and the influence of CO₂-, electricity- and solar heat supply cost

The lowest line in **Figure 6**, considering the cost for CO₂ supply, electricity consumption and solar heat supply for methanol distillation, hits the “threshold” of 700,000 € at PCE of roundabout 12%. Furthermore, the cost assumptions for electricity and solar heat supply are rather optimistic/low. These circumstances imply, that the photocatalytic reactor field shall have a PCE above 12% to support itself in economically respect. Or, on the other hand, it has to be estimated, if the reactor field can be build for maximum 700,000 €/hectare.

350 €/t methanol is close to the Free On Board price of methanol in the harbour of Rotterdam in Netherlands in the first Quarter 2012 [16]. To demonstrate the effect of lower methanol price, **Figure 7** shows the hectare specific net present value like in **Figure 6**, with methanol price set to 250 €/t. The “threshold” of 700,000 € per hectare is now exceeded at roundabout 18% PCE. Compared to a methanol price of 350 €/t the net present value drops substantially.

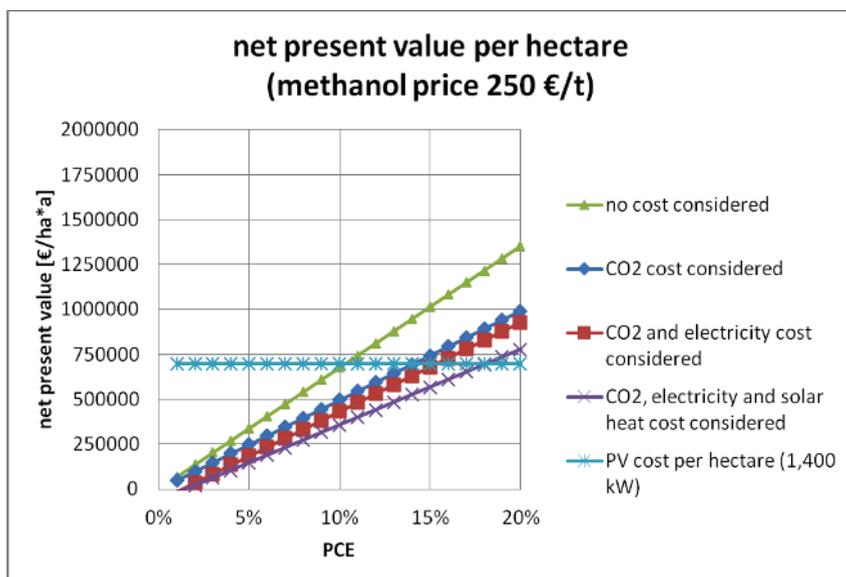


Figure 7: Hectare specific net present value calculated with methanol price of 250 €/t in dependence on PCE and the influence of CO₂ supply, electricity- and solar heat supply cost

3 Conclusion

In this paper, the energy and greenhouse gas balance of photocatalytic CO₂ conversion to methanol was evaluated. The process chain included the CO₂ capture and liquefaction in coal fired power stations, CO₂ transport via far distance of 3,743 km from Germany to Algeria, photocatalytic conversion and methanol purification to fuel grade for application in combustion engines in the traffic sector.

It was figured out, that the CO₂ capturing and liquefaction step is a very energy intensive step in the process chain. CO₂ pipeline transport to Algeria is highly energy efficient and does nearly not influence the energy- and greenhouse gas balance.

Compared to the today's situation of power stations without CO₂ capture and oil based gasoline fuelled cars, the "Solar2Fuel" system with CO₂ capture, liquefaction, transport and photocatalytic conversion to methanol can decrease the greenhouse gas emissions by about -39.6%. This is even better than the relative reduction of carbon capture and storage in underground formations where cars would be further fuelled with oil based gasoline. The CCS case would reduce the greenhouse gas emissions about -32.2% compared to the today's situation. To compete with the CCS case, the CO₂ conversion to methanol in the "Solar2Fuel" system needs to exceed 77%.

In terms of primary energy consumption, the "Solar2Fuel" system can offer a maximum reduction of -49.0% compared to today's situation.

It was shown, that CO₂ supply price from coal fired power stations inclusive transportation and overnight storage in Algeria is about 46.0 €/t. In case of a methanol value of 350 €/t, economic evaluations showed, that a hectare specific net present value of a photocatalytic reactor field can exceed a defined threshold of 700,000 €, if the photo conversion efficiency exceeds 12%, which means that 12% of the solar energy is stored in methanol in terms of higher heating value.

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