

Application of Nano-Membranes in CO₂ Bioconversion

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ABSTRACT

CO₂ emissions from the power stations become a commodity if used for intensification of photosynthesis in various bio-systems- greenhouses, crops cultivated in free air, growing trees and especially algae ponds. Application of nano-membranes permits to supply to the above biosystems flue gases with increased CO₂ concentration. Few examples of such application are presented as well as a first-approach evaluation of expected benefits.

1. INTRODUCTION

The power stations CO₂ emissions mitigation is commonly understood as a chain of three main links: capture, i.e. separation of the carbon dioxide from the flue gases, transportation and storage. According to the common logic of the abovementioned chain, after separation from flue gases carbon dioxide has to be liquefied by compression (to decrease dramatically its volume) and so it can be transported to the sinking area and pumped in an underground sink.

For separation process the mostly known technology is chemical absorption by aqueous amines or other substances. This process is widely used in chemical industry, when the goal is CO₂ production. These substances dissolve the CO₂, separating it from other flue gas components. The solution is then regenerated by heating and the released carbon dioxide is processed for transportation and storage.

The main disadvantage of such absorption-stripping process is the high energy consumption, mainly of the thermal one for stripping. Major efforts are made now for development of new generation of absorbents which will permit a dramatic reduction of the needed heat, but they still are in the experiments stage.

Another technology of CO₂ separation, using gas separation nano-membranes, seems to be more promising, mainly due to lower energy requirements and is intensively developed in various countries. The 26 European research and industrial organizations united their efforts under umbrella of the NanoGLOWA Integrated Project, supported by EU [1]. The project goal, approved

by EU, is development and testing of few different separation membranes, based on various novel materials. Various system configurations (pressure on the feed, vacuum on the permeate side, or combination of both of them) have to be considered with the aim to “increase of the CO₂ concentration up to 95%, at a price of ~ 20 euro per ton CO₂.”

Meantime there is a field of bioconversion of CO₂ which can contribute to mitigation of the CO₂ emissions by using it in the photosynthetic process which is one of the mostly feasible ways for mitigation of CO₂ emissions.

Photosynthesis in green plants and microorganisms achieves CO₂ fixation on a global scale. The incorporation of CO₂ into the biosphere by the photosynthetic action of plants and microorganisms has been estimated to an amount of about 10¹¹ tons CO₂ per year, which is converted together with solar energy into biomass.

Under field conditions, the efficiency of solar energy conversion in plant production is not high - the global average efficiency has been estimated at 0.15%. However, under optimal growth conditions it is much higher, reaching sometimes even 5 to 6%. The optimal conditions for efficient photosynthesis can be created in both closed (like greenhouses) and open areas and so to provide absorption of significant quantities of CO₂.

On the other hand, cultivation of plants in CO₂ enriched atmosphere provides, besides CO₂ sequestration, economic benefits due to improvement in both the yield and quality of the plants and decrease of required irrigation water. Large scale application of such enrichment is limited mainly by CO₂ costs.

Flue gases of power stations supplied through pipelines could be a cheap source of CO₂, but their use is limited by presence of harmful pollutants, like SO₂. The second shortcoming of this CO₂ way of supply is its relatively low content in the overall flue gases. This shortcoming implies large gas deliveries for small CO₂ usage. It means expensive pipes and wasted energy for the transportation, proportional to the cultivation site distance.

State of the art of the nano-membranes technology permits yet an increase of the CO₂ content up to ~ 60% (instead of the typical 12-14 % in the stack of a coal fired power station equipped with desulfurization system) in relatively simple and easy way, in one-stage process. Thus, there are technical means to supply flue gases (after a pretreatment and passing through nano-membranes system) to bio-systems (like greenhouses, algae growing ponds and so on), providing both CO₂ mitigation and economic benefits. Although this idea can not be panacea, it is simple and feasible in many places; let us mark that it can be especially efficient if applied for growing of terrestrial or aqueous biomass for bio-fuel production. Stack gases with an increased content of CO₂ can be

supplied through polyethylene pipes to rather big sites (thousands of hectares) at costs strongly competitive with the above marked chain “separation-transportation-storage.”

In the chapters hereafter, we will try to list fields where the bioconversion projects can be performed (Chapter 2), describe our experiments with coal fired power station flue gases (Chapter 3), and try to show where nano- membranes application has advantages (Chapter 4).

2. BIOCONVERSION WAYS

CO₂ bioconversion occurs all the day long; various plants absorb CO₂ from the atmosphere and convert it into oxygen and carbon due to solar action. Hereafter we refer not to this natural process, but to cases (existing or possible) of intensified photosynthesis by CO₂ enrichment, i.e. in the atmosphere with a higher content of CO₂. We try to show feasibility of use of flue gases of power stations for this aim.

2.1.ENRICHMENT OF GREENHOUSES AND FREE AIR CULTIVATION

Enrichment of the greenhouse atmosphere by CO₂ proved to be very useful, providing significant increase of the yield, improved quality of plants and economy in water consumption and is widely applied in many countries. For most crops, CO₂ concentration increases up to 700-1000 ppm over a long period and increases growth and production by 20-30% [2].

In countries with high solar activity, like Israel, the enrichment of greenhouses with CO₂ is especially efficient, because of the large number of sunny days in winter. Research of the Agriculture Research Organization (Israel) showed ability to increase tomato yields by 30% (or by 100% if combined with heating), and melon yields by 100% and so on, but high prices for CO₂ (400 -800 € per ton) limit its application [3].

From the point of view of the potential CO₂ mitigation, enrichment of greenhouses has been evaluated as follows (on the example of Israel). There are 5 400 hectares of greenhouses in Israel, 2 000 of them ready for immediate implementation of CO₂ enrichment. The average rate of efficient enrichment in CO₂ is ~6 g/m²/hour, or 60 kg/hectare/hour. In conditions of high solar radiation and low cost of CO₂ from flue gases, one can assume that enrichment will be efficient during 2600 hours per year. Thus, the project development can provide- potentially- mitigation of ~ 310 000 ton CO₂ per year ~ 10% of annual CO₂ emissions from a 550 MW coal fired unit.

It is important to emphasize that increased CO₂ concentration in the greenhouse environment provides essential (20-40%) economy of used water. Water consumption in greenhouses is 30-60 m³/hectare/day. Thus, water economy potential per year can be evaluated as 15 million m³, if average rates are considered. Considering that water is produced by desalination with average

power consumption $\sim 4 \text{ kWh/m}^3$, another 50 000 ton of the CO_2 emissions can be avoided. Notwithstanding the above advantages, large scale application is limited by high prices of CO_2 .

In the world, CO_2 for enrichment is normally obtained in liquid form or produced directly in the greenhouse atmosphere by burning hydrocarbon fuels such as natural gas, LPG propane, and premium kerosene (paraffin) which all contain low and acceptable levels of sulfur oxides. Some countries (Britain, Netherlands and others) encourage growers in these countries to practice CO_2 enrichment by ducting flue-gases from centralized, gas-fired boiler installations into their greenhouses.

Generating CO_2 from hydrocarbon fuels can give rise to several gaseous air pollutants that are potentially damaging for crop production. Although low-sulfur fuels are used and other special measures are undertaken, like design of low- NO_x burners there is still risk of incurring losses in yield due to air pollutants [4].

Free-air CO_2 enrichment (FACE) experiments with many species of crops and other vegetation have been and are being conducted in many countries. In USA, for example, long term research program FACT1 concerning forests is supported by US DOE [5-6].

The purpose of the studies is to determine the potential long-term response of vegetation to rising concentrations of atmospheric carbon dioxide, to locate a possible sink for carbon dioxide, to determine the factors that enhance and/or limit that sink, and so on.

As the main goal was to predict growing in expected future increased CO_2 atmosphere, thus in the main part of experiments CO_2 concentration was above ambient by 100-200 ppm, i.e. below 700-1000 ppm usual for the greenhouses. As expected, the elevated CO_2 increased photosynthesis and biomass production, especially provided better development of the roots, and greatly improved water-use efficiency in all the crops. There is sufficient data for planning of large scale enrichment projects which can serve mitigation of CO_2 , where a big part of the costs is compensated by increased yield and quality of crops and reduced irrigation water consumption.

2.2 CULTIVATION OF MICRO ALGAE

Photosynthesis in algae is much more intensive than in terrestrial plants. Thus, microalgae cultures require a concentrated CO_2 source, such as that available from power plant flue gases. There are many efforts to apply intensive microalgae cultivation with the aim to mitigate CO_2 power plants [7-8]. The grown microalgae can be used for production of high-value chemicals, nutritional products, in wastewater treatment and so on. In the last years microalgae are being studied as a promising source for vegetable oils suitable as feedstocks for bio-fuels. The advantage of microalgae is their potential to achieve much higher productivities than conventional oil crops, in

part due to their continuous production process and overall high oil content. Let us emphasize, that one can produce from algae up to 30 toe (ton oil equivalent, ~42 GJ) per hectare as compared to 0.5 - 1.5 (up to theoretical 5) toe from terrestrial plants. Moreover, no sweet water and no fertile lands are needed. Microalgae can use waste, sea, and brackish waters and land resources that are unsuitable for crop agriculture; algae fuels contain no sulfur, are non-toxic. From the point of view of the CO₂ mitigation, the best known algae growing projects show productivity of about 25-30 g per sq meter per day; this means 50-60 g net CO₂ absorption; besides this value, certain quantity of CO₂ will be lost dissipated in atmosphere and absorbed by seawater.

To make large scale algae biofuels production economic some more R&D efforts are needed to develop hardy, high oil content, high productivity algal strains that can be stably cultivated and easily harvested, and to improve methods both for growing and oil extraction.

3. EXPERIMENTS ON BIOCONVERSION OF THE RUTHENBERG POWER STATION

CO₂ EMISSIONS Few experiments with the goal to prove the feasibility of bioconversion of CO₂ contained in a coal fired power station flue gases were performed at the Israel Electric Co (IEC) Ruthenberg Power Station (RPS) units .

3.1 GREENHOUSES ENRICHMENT

Flue gases of LPG or kerosene are widely used for CO₂ enrichment of greenhouses. Sometimes flue gases of natural gas fired gas turbines are used. Flue gases of the coal fired power units containing sulfur oxides and other pollutants are not used at all, even if the power unit is equipped with FGD system. The main reason is that existing FGD systems do not provide sufficient removal of sulfur dioxide.

It is important to emphasize that requirements against contamination by the flue gases used in greenhouses are much more severe than the general requirements of environmental legislation.

An R&D project [9], dealing with the coal fired power station flue gases use for photosynthesis intensification, has been accomplished in 2002-3. The project aim was to demonstrate that flue gases from coal-fired power plants, can be treated and used in greenhouses for CO₂ enrichment. The RPS is equipped with the FGD systems. The measured SO₂ content in the flue gases is ~ 70 - 180 mg/dNm³.

Table 1. Maximal acceptable concentrations for humans and plants of some components of the flue gases (vpm).

Gas	Humans	Plants	Plants long term exposure
CO ₂	5000	4550	1600

CO	47	100	
SO ₂	3.5	0.1	0.015
H ₂ S	10.5	0.01	
C ₂ H ₄	5.0	0.01	0.02
NO	5.1	0.5	0.25
NO ₂	5.0	1	0.1

For greenhouse enrichment one has to supply 6 g CO₂ per 1 m² per hour. Considering 15.5 % CO₂ concentration (dry basis) in flue gases, this means 0.02 dNm³ flue gases per 1 m²/hour. SO₂ concentration will be 0.1- 0.2 vpm. This value is unacceptable for long time exposure and has to be reduced by ~ 5 times. This can be reached in two ways:

- supplementary scrubbing of the gases stream to be supplied to the greenhouse
- increase of the CO₂ concentration by nano-membrane, reducing in this way total of gases supplied; thus, the SO₂ content will decrease, at least in parallel.

In order to show the feasibility of the enrichment we supplied flue gases with increased concentration of CO₂ in cylinders. For the concentration increase we used the Carbon Molecular Sieve Membrane (CMSM) technology, kindly provided by the firm "Carbon Membranes Ltd" (CML) (Israel) [10]. CML gas separation systems are based on original hollow-fibre carbon molecular sieve technology. Molecular sieving is a mechanism whereby different molecules are separated because of their different size. For the porous carbon nano-membrane this is the main (but not always the only) separation mechanism. The separation module which has been used in our experiments consisted of a large number of fibres ~ 10,000 within a stainless steel shell. The module was designed to ensure maximum circulation of the feed gas to optimise the separation process, along with durability to withstand field conditions.

The results described below were obtained with one-end-opened type pilot module, composed of approximately 10,000 carbon hollow-fibres, having an active separation area of 3.4 m².

Fig. 1 shows a scheme of an experimental system mounted at RPS for preliminary treatment of flue gases, increasing of CO₂ concentration and compression in cylinders.

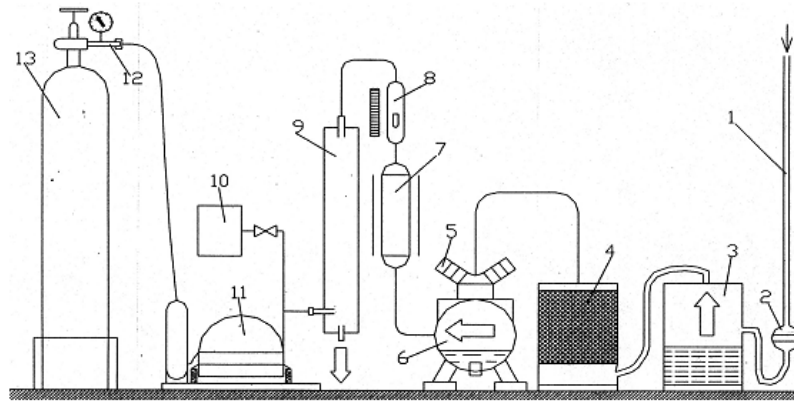


Fig. 1. Experimental System for Filling Cylinders with Flue Gas with Increased CO₂ Concentration.

The wet flue gases after FGD system were extracted from the stack, from an opening at 150 m height, normally used for sampling of the gases. Vapour condensation in the pipe caused partial removal of SO₂ and NO_x. Drained and scrubbed flue gases from condensation tank 3, are sucked by compressor 5 through activated carbon filter 4 and supplied to receiver 6. An additional amount of moisture is removed. The pressure in the receiver is about 8 bars. The moisture content in gases in 7 is absorbed by granules of silica gel. The flow of gases is indicated by flow meter 8. Further drained and scrubbed flue gases pass to the carbon nano-membrane 9 for CO₂ concentration increase. The pressure drop of the gases at carbon nano-membrane is about 6 bar. Gas composition was examined using the gas analyzer 10. The scrubbed, drained and concentrated flue gases are compressed into the cylinder 13 by the compressor 11, able to create pressure up to 70 bar.

Table 2. Concentration of CO₂ and pollutants in flue gases after FGD and supplementary treatment (example).

CO ₂ [in %]	CO [ppm]	S O 2 [p p m]	N O [pp m]	Note
8.6-9.9 %	70-80	20-25	40-50	Before CMSM
44-46 %	40-50	3-5	4-6	After CMSM

Series of experiments on the influence of the atmosphere enrichment by flue gases with the above composition on various plants growing have been performed. Prescribed CO₂ concentration (1000-1300 ppm), in the seedling cultivation area, has been maintained by compressed gases supply control.

In the first series of experiments, germination of the roots system with and without enrichment have been compared (Fig.2). CO₂ was maintained at 1000 ppm. The tomatoes Fig 2 (a) were grafted and placed in CO₂ enriched atmosphere during six days. The watermelon was grafted and treated by the CO₂ enriched atmosphere during 7 days Fig. 2 (b) and (c).

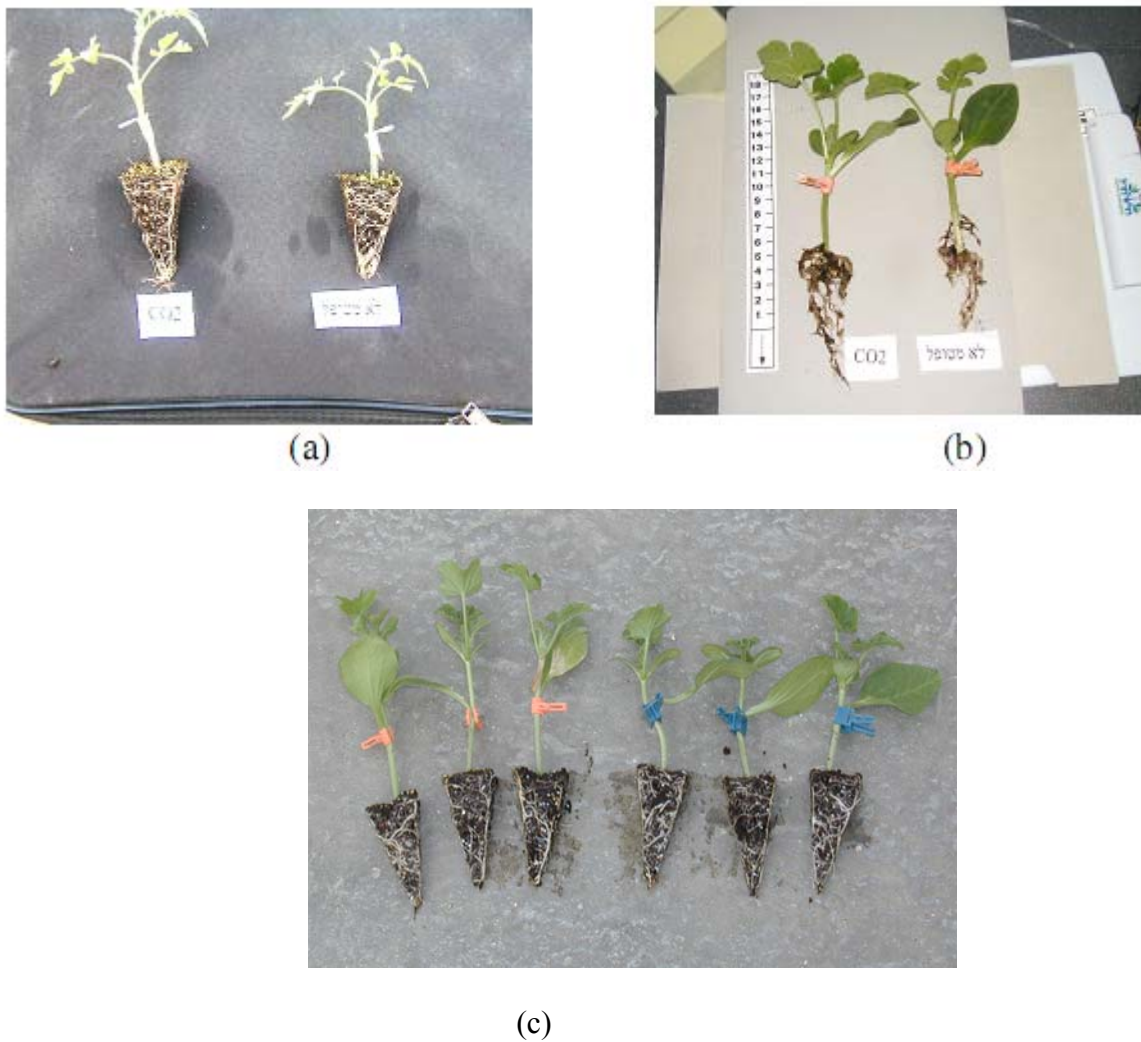


Fig. 2. The comparison between CO₂ treated and control plants.

Dry mass of the treated plant root was higher by 15% average.

Statistical analysis of experiments with tomato sort "Crimson" treated with flue gases and control showed (reliability not less than 90%) changes of weight of seedling (grafting, cutting and rootless). The increase in an additional weight of inoculation as a result of influence of flue

gases in series of experiments reached approximately 22% (with reliability 96 %); increase of an additional weight of the rootlet was 40.4 % (with reliability 98 %).

3.2. ENRICHMENT OF THE ALGAE PONDS

A prototype algae production plant with about 1,000 m² of ponds has been established and is operated in the RPS by Seambiotic Ltd in collaboration with the IEC with the aim to demonstrate the feasibility of cultivation of marine microalgae in the open seawater ponds enriched by flue gases of the coal fired power station and to develop efficient cultivation techniques which will permit large scale algae cultivation at cost ~ 0.2- 0.25 euro per kg dry mass.

Algae are cultivated in four pairs of ponds (with area of 5 m², 20 m², 100 m² and 300 m²). Flue gases are supplied to the ponds from the stack, through a supplementary treatment system and KREAL diffusers. Productivity of the algae cultivation is higher than in similar ponds enriched with pure CO₂ from the tank.

Table 3. Average composition of flue gases at RPS.

CO ₂ , %, Volumetric (wet)	13
N ₂ %, Volumetric (wet)	69
O ₂ %, Volumetric (wet)	5
H ₂ O %, Volumetric	13
CO, mg/dNm ³	40
NO _x , mg/dNm ³	450
SO ₂ , mg/dNm ³	120
Solid particles, mg/dNm ³	20 - 40



Fig. 3. Experimental Cultivation of Marine Microalgae in the RPS

Flue gases are a cheap and unlimited source of CO_2 , but its low concentration and difficulty to be liquefied, limits their application for algae ponds. The necessity to supply and to disperse big volumes of the gases; if the ponds are situated at a distance from the power station stack, the advantages of the cheap CO_2 source use should be reconsidered. Let us emphasize that for large scale algae cultivation the CO_2 supply issue is crucial. This problem, we believe, can be solved by application of the novel nano-membrane technologies, providing dramatic increase of CO_2 concentration.

First, small-scale tests of applicability of the gas separation membranes for algae growing have been performed in 2007-2008. The goal was to show that flue gases with CO_2 concentration by nano-membranes provide the same cultivation efficiency, as the flue gases.

The membrane has been connected according to the diagram below (Fig. 4). The accurate lab scale tests showed no difference between algae cultivated with gases "as is" or gases with increased CO_2 content.

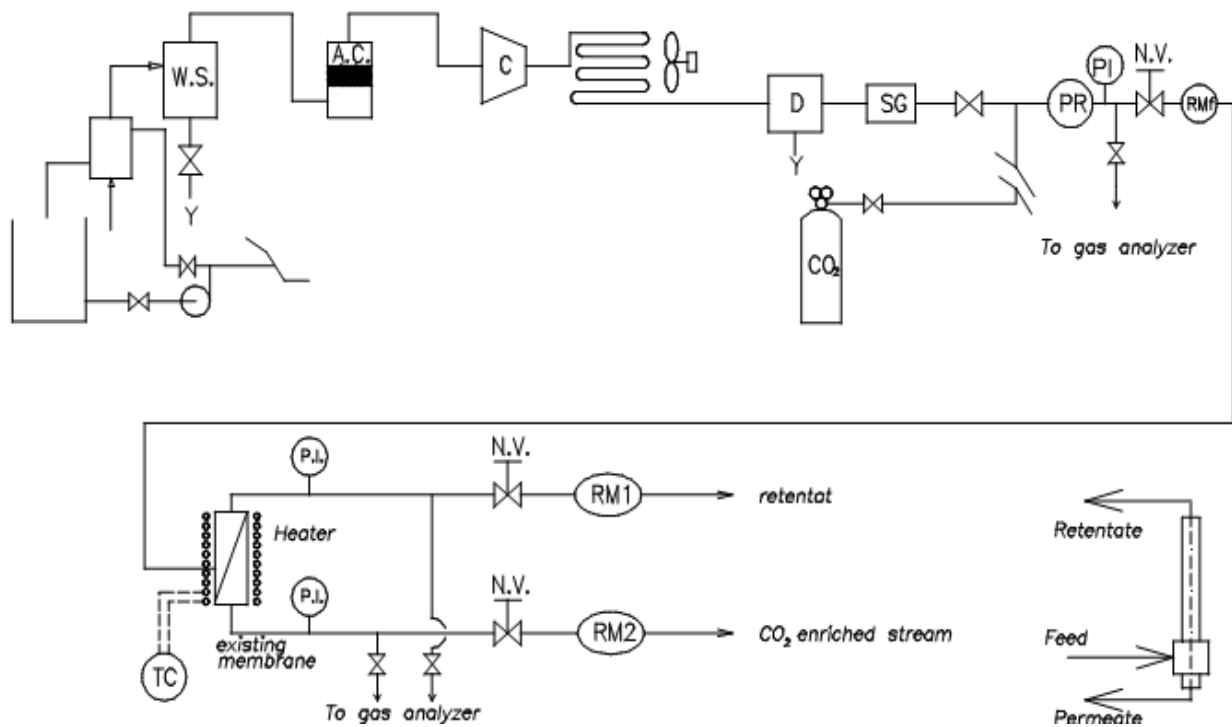


Fig. 4. Nano-Membrane Test Plant for CO_2 Enriched Stream.

The next experiments have been done using the commercially available gas separation membrane produced by Parker Filtration & Separation B.V. (Fig. 5), which are asymmetric hollow membranes made from high molecular weight poly(phenylene oxide), PPO. PPO is a semi-crystalline polymer with a high intrinsic gas permeability but moderate selectivity. However, the combination of the high intrinsic gas permeability of PPO with the industrial hollow fiber

production capability of producing asymmetric hollow fiber membranes, in one step, with an outside skin layer thickness of less than 40 nm provides high permeability of the nano-membrane developed in the framework of the NanoGLOWA project by Parker Ltd (Fig. 5).



Fig. 5. Parker Nano-Membrane Installed in the Test Rig in the Algae Pilot Plant.

In order to optimize the separation of CO₂ a vacuum pump on the permeate line has been added to improve the driving force and enhance the separation performance of the membrane module, as shown in the diagram (Fig. 6); (three membranes shown in the diagram are optional).

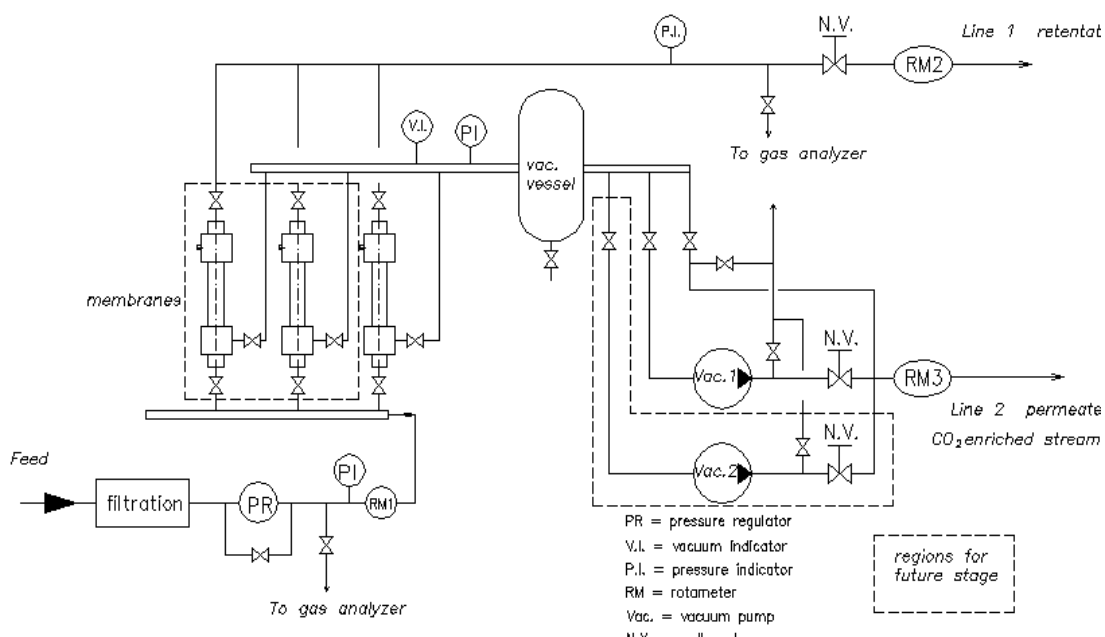


Fig. 6. Membrane Test Rig with Compressor and Vacuum Pump.

In the first test with the Parker module the gas separation performance was tested under different conditions by applying flue gasses from the Power plant as shown in Figure 6. The module was tested with bore side and shell side feed, resulting in the best performance using a pressurized bore side feed with an additional vacuum on the permeate side. In one single stage the concentration of CO₂ could be enhanced from ca. 12% to over 30% with a CO₂ recovery rate of ca. 74% using a low inlet pressure of 1.5 bar(g) and a vacuum on the permeate side of 0.5 bar(a). By using a lower enrichment as target as e.g. 24% CO₂, the CO₂ recovery was increased to over 90%.

These first results indicate that with a simple 2 stage system, purities of more than 60% with high CO₂ recovery rates could be achieved.

Membrane module developed by NTNU is also prepared for testing (Fig. 7).



Fig.7. NTNU Nano-Membrane Module Installed in the Test Rig.

Now parallel tests of algae growing with enrichment with flue gases "as is" and with 24% CO₂ are performed; in the both cases PH = 7 is maintained.



4. COMPARISON OF PURE CO₂ AND FLUE GASES WITH INCREASED CO₂ CONCENTRATION SUPPLY.

An example demonstrating advantages of nano-membranes application for supply of CO₂ to bioconversion site is shown hereafter.

Assume a large marine algae cultivation farm at certain distance from the coal fired power station stack with CO₂ consumption 100 ton per hour and let us compare three options of supply of CO₂:

- 1) pure CO₂ produced using MEA process (not liquefied);
- 2) flue gases with CO₂ content increased using nano-membranes;
- 3) flue gases – as outcome of the stack.

Let us note, that the first two options include processing of gases with the aim to produce the pure CO₂ or flue gases with increased CO₂ content, which has both fixed costs (mainly investments) and variable costs (mainly energy consumption). In our evaluations we will refer mainly to the last component. As to the supply of the flue gases (option 3) the first evaluations showed that it is not feasible if distance is more than several km.

4.1. SUPPLY OF PURE CO₂.

Assume CO₂ produced in the power station using the chemical process of absorption by MEA or other similar substance solution. The process includes stages of absorption and recovery of CO₂.

Average heat consumption is 1.7 ton of low potential steam for recovery of one ton CO₂; the last research show that this value can be reduced down to 1.5 ton. If such quantity of steam is extracted from the steam turbine at low pressure ~ 6 ata (and if such extraction is technically possible) lost

electric capacity is in this case ~ 30 MW for production of 100 ton CO₂. Electricity consumption for CO₂ production is in the range of 50 kWh per one ton CO₂, or 5 MWh per 100 t; i.e. total electricity per 100 ton CO₂ is 35 MWh.

For the supply of 100 ton CO₂ per hour through a plastic pipe with internal diameter 2 m at a distance of 100 km, for example, another ~ 1 MW is needed, thus total electricity (consumption and lost) is ~ 36 -37 MW as function of distance (Fig. 8).

4.2. SUPPLY OF FLUE GASES WITH INCREASED CONCENTRATION

In the framework of the above assumption (only a part of the total CO₂ in flue gases has to be separated) a nano-membranes battery can be used to increase concentration up to 60 %. The CO₂ permeation through nano-membranes will be provided by both positive pressure on the feed side (2 ata) and vacuum pump creating absolute pressure 0.25 bar. Electricity consumption will be ~ 24 MW (the evaluation has been kindly provided by Prof. May-Britt Hagg from Norwegian University of Science and Technology in Trondheim [14]). For transportation of the flue gases through the same 2 m diameter pipe to 100 km distance electricity consumption is about 3 MW. Thus, total electricity consumption is ~ 27 MW for this case.

Comparison of the above options is shown in Fig. 8.

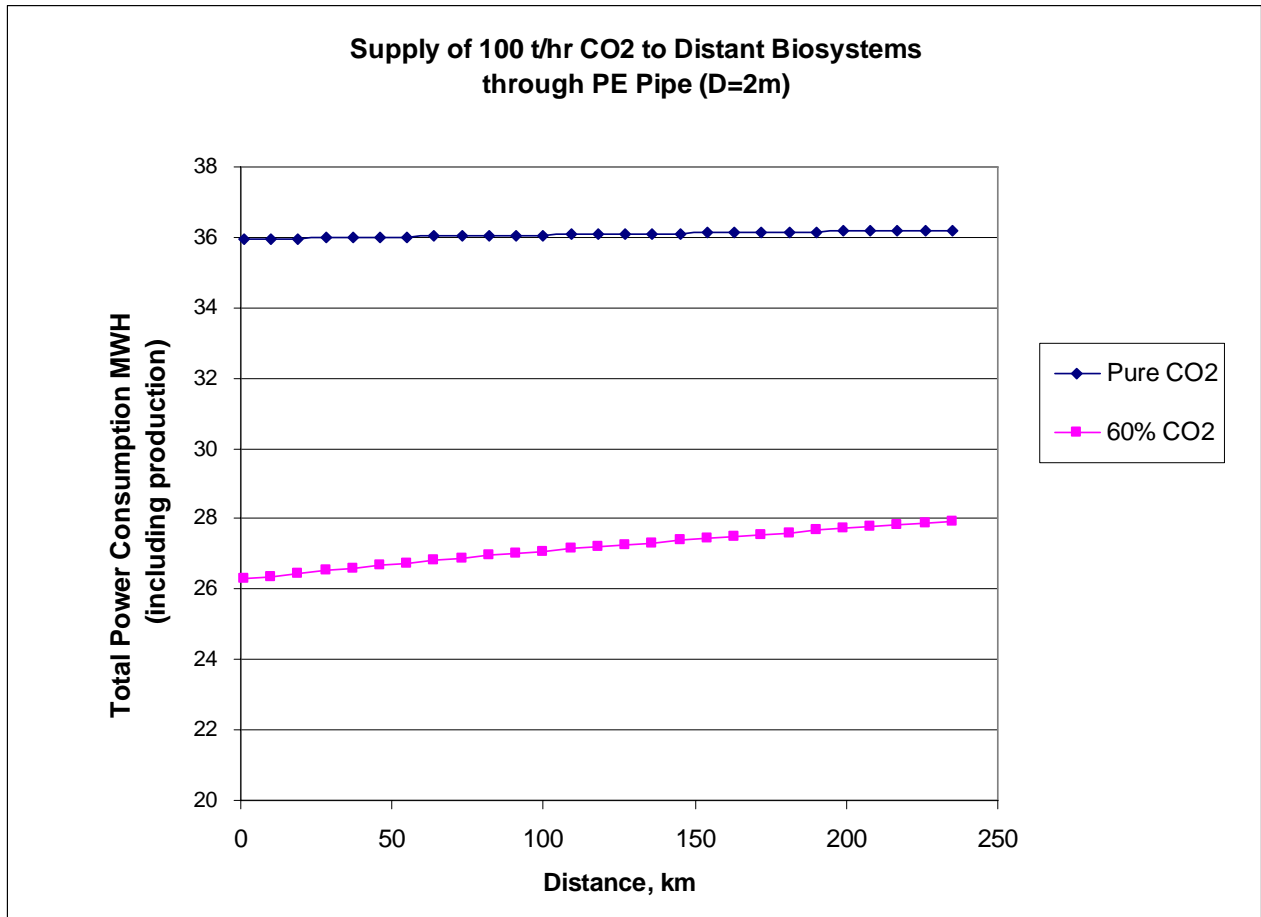


Fig. 8. Comparison of Electric Power Consumption

Comparison of the two options refers only to power consumption; for the pure CO₂ option lost electric energy generation is included, as well as power consumption for the pure CO₂ production and transport. In the flue gases with 60% CO₂ energy consumption, we included energy consumption for both vacuum pump and for transport.

We did not include in the comparison total investment needed for the both cases, but on the qualitative level one can state that investments in membranes facility with the aim to increase CO₂ concentration are lower or at least of the same magnitude as investments in CO₂ plant.

4.3. SUPPLY OF FLUE GASES WITH 13% CO₂ CONCENTRATION - AS OUTCOME OF THE STACK

Supply of the flue gases (option 3); the first evaluations showed that it is no feasible if distance is more than several km, besides the fact that for enrichment of terrestrial crops a supplementary scrubbing is necessary.

5. CONCLUSIONS

We believe, that profitable projects of bioconversion of carbon dioxide emitted by power plants are feasible and we put efforts to prove it. Such projects have potential to contribute to mitigation of the emissions with no hard load on the power stations because a big part of cost can be offset by revenues.

Application of nanomembranes for increase of the CO₂ concentration in flue gases of coal fired power stations has potential of essential decrease in the costs. Concept of such projects can be developed in various countries, with regard to the cultivated plants, existing vegetation and so on, i.e. the biosystems which can be "hosts" of the CO₂, from one side and to the location of the industries generating CO₂ (like power plants) from the other. Industrial pilot projects have to be erected as the next step to develop this idea.

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