Technical paper

Potential Solutions for CO_2-capturing Technologies in the Slovenian Context

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Dedicated to Prof. Dr. Gorazd Vesnaver on the occasion of his 70th birthday

Abstract

Human activities have caused an enormous rise of the CO_2 concentration in the Earth's atmosphere over the past 200 years. In order to alleviate this problem, the threats to and the concerns of the international community need to be converted into economic opportunities for national economies, which shall develop and utilize technological opportunities rather than simply accepting international obligations to reduce CO_2 emissions. In the article we analyze technological possibilities in the Slovenian context as possible opportunities for promoting sustainable development based on regional, renewable resources. Beginning with an analysis of the amine process for CO_2 concentration and its possibilities, we continue with CO_2 chemistry examples, like the precipitation of calcium carbonate from Ca^{++} sources like lime or fly ash. Through the concept of product engineering we emphasize the need for a stepwise realization from the laboratory to a pilot plant and then to the industrial scale. The growth of biomass through forestry or algae production can provide an additional CO_2 sink. However, for an efficient technical solution and implementation a close working relationship between biologists and engineers is required.

Keywords: CO_2 minimization, technological opportunities, amine concentration, CO_2 chemistry, forestry, algae production, fly ash CO_2 absorption.

1. Introduction

The governments of the EU Member States have prepared a variety of documents relating to CO_2 emissions. In a Slovenian implementation report[1] some conclusions were drawn to point the way to an overall solution: "a more consistent implementation of the reform programme would be required," which is a typical administrative view or reaction to the situation. In order to find new possibilities we have tried to identify some technological opportunities that could serve as a basis for business opportunities. The identification and presentation of some technological opportunities could be an important way to support the dissemination of new ideas to the business sector so as to become drivers for new developments and investment cycles.

The identification of technological opportunities within national and local boundaries needs to be made with knowledge of the local circumstances, i.e., relations and elements have the main influences on the base premises of the technological opportunities. With such a background, some promising technological opportunities have been identified and investigated as a basis for R&D efforts, but all are strongly connected to environmental issues like CO_2 capturing and support to sustainable development models.

At this point the new research programs are based mostly on a "pull" strategy. Following the concept of the so-called "research development market", some great opportunities exist to introduce BAT technologies so as to increase the limitations of environmental protection, without neglecting sustainable development.

The frightening increase in the concentration of CO_2 in the atmosphere that we are facing – Figure 1, which has the potential to cause natural catastrophes and climate changes, is putting serious pressure on decision makers to act globally, for example, at the Kyoto, Cancun and Durban international conferences on climate changes.

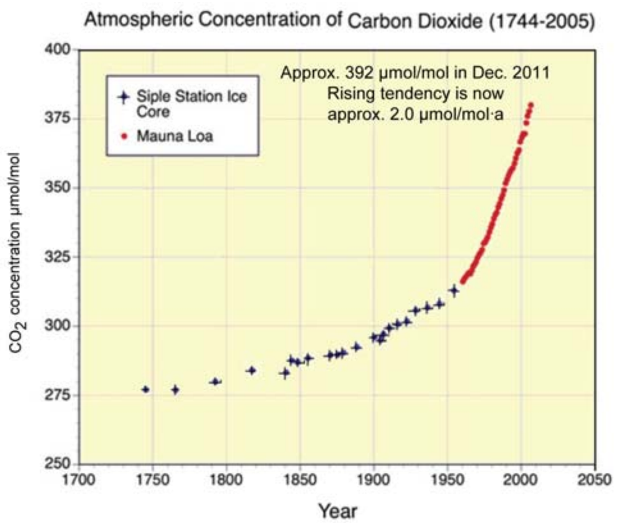

Figure 1: Atmospheric Concentration of Carbon Dioxide since 1744 to the present day

CO_2-related policies are calling for a reduction in CO_2 emissions, CO_2 capturing, trading, the shutting down of major CO_2 emission locations and support for low-carbon energy production.

Taking into account all the circumstances and the rapidly changing framework for the development and implementation of new technologies at national levels, some different concepts are appearing around the world. Typical examples include wind-power plants in Germany, the optimization of nuclear power plants in France and the renovation of coal power plants in Poland.[2] The important lessons here are that countries try to discover real and pragmatic options that are acceptable for both the local environment and the national economy. In this context, we have collected and reviewed some technological options that already exist in Slovenia and that have the potential to become important parts of the Slovenian effort to shift to a low-carbon society and also to serve as a trigger for sustainable developments in the regions.

2. Forestry

There is a great deal of talk about employing forestry as the basis for an important carbon-sink reservoir. This is especially important for Slovenia, where forest occupies about 60% of the country's area.[4] Successful forestry policy and practice over the past 50 years could be extended to the intentional increase of the area of the forest to support carbon sink. The balance between emissions and the CO_2 sink is positive and could reach 3–6 t of CO_2 absorption per hectare per year (3–6 $t/ha \cdot a$). Converting this into the Slovenian context and mass units, the data shows that wood increment is at a level of 6.78 $m^3/ha \cdot a$, which,[4,5] according to calculations, could be about 3 $t/ha \cdot a$ of wood in mass units.

With the activation of more than hundred hectares or even 1000 hectares of land for forestry, significant quantities of CO_2 could be stored. Also, such activation could open up possibilities to utilize renewable biomass for the substitution of heating systems based on fossil fuels. However, even if the economics of forestry is promising, the initiative needs to be connected with the obligations of fossil-fuel electricity producers (as the largest CO_2 emitters) and national targets in the environmental and social areas. In times of crisis it is vital to reconsider the loose connections between environmental obligations and national policy on sustainable development.

Besides conventional forestry, planted forests could also be an important sink for CO_2. If planted on cleared, agricultural land, plants can help to remove 5–30 $t/ha \cdot a$ of CO_2 from the atmosphere.[6]

3. CO_2 capturing

The concentration process of the flue gases, where the volumetric concentration of CO_2 is in the range from 8–12%, can be performed with one of the classic pressure-swing processes (PSA), i.e., the amine process. The problems associated with adapting the process from conventional applications, like H_2 production from CH_4, have been intensively investigated.[7] The classic amine process is presented in Figure 2 with the assigned operating points. The process is originally designed for H_2 production, where concentrated CO_2 is a side product, used as a pure gas in various industries, or subject to potential, but still questionable, solutions involving geological or oceanic storage.[8]

The feed gas consists of hydrogen and CO_2 from the methane steam-reforming process:

$$CH_4 + 2H_2O \rightarrow 4H_2 + CO_2 \quad (1)$$

The CO_2 reacts with amines in pressure–temperature swing absorption/desorption cycle process according to the summary reaction:

$$CO_2 + R_2NCH_3 + H_2O \leftrightarrow R_2NCH_4^+ + HCO_3^- \quad (2)$$

Problem of the classic amine process design, which comes from the differences in the input gases, is presented in Table 1. The flue gases are always under normal pressu-

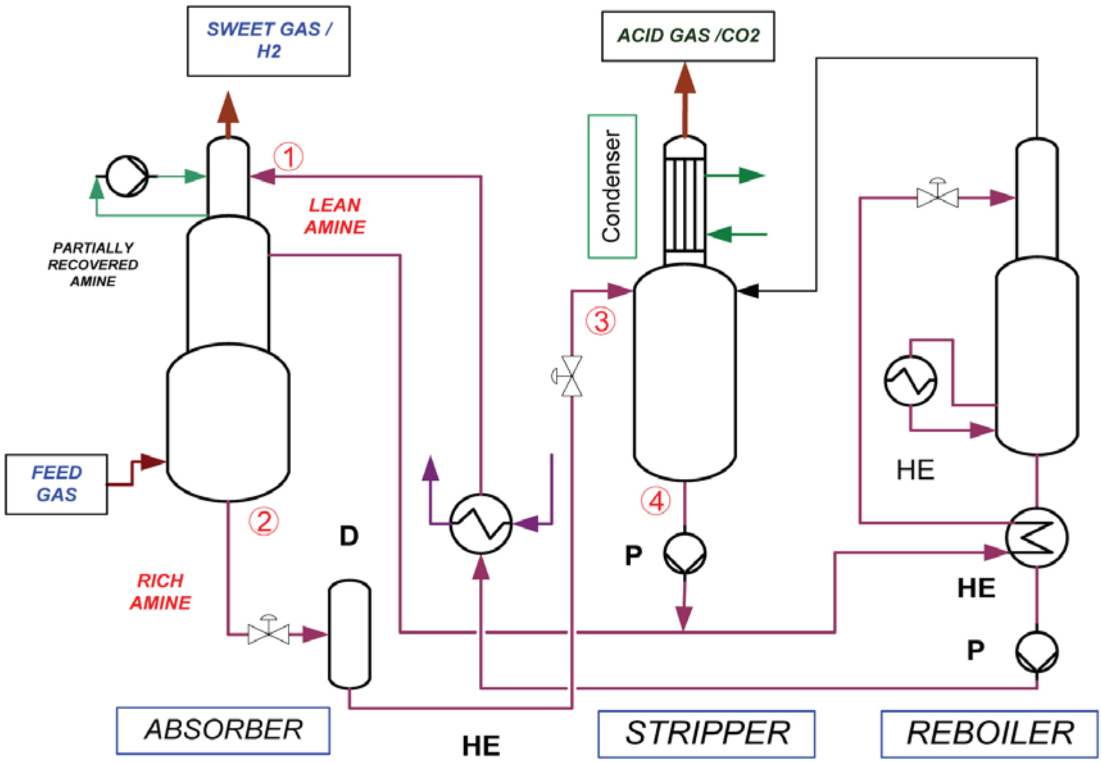

Figure 2: PSA amine absorption process unit for CO_2 concentration

re, compared to natural-gas reforming, where the pressure could be from 10 to 100 times higher. The biggest differences are in the gas flow, where the ratio is about 1000, and in the CO_2 concentration, which is 1:10. The quantities of flue gases are enormous, but the CO_2 concentration is always relatively low. Flue gases contain about 10 times more oxygen than a reformed mixture from natural gases. A major consequence of these differences, species concentration and the amount of gases is an enormous rise of the process energy costs. The ratio of the energy cost to capital cost is about 10 times higher for flue gases than for a natural-gas reforming mixture treatment – Table 1. The high energy cost of the CO_2 concentration from flue gases has given rise to some new research proposals that might provide new solutions[8] – some new commercial solutions have been announced.[9]

Table 1: Comparison of CO_2 capturing from selected streams

	Flue Gas	Reformed natural gas
P_{tot} /bar	1	10–100
Gas Flow	1.7–4	1.7–170
	M(m^3)/h	k(m^3)/h
P CO2 /bar	0.1	1 to 10
PO2 /bar	0.02–0.1	0–0.01
ratio: Energy cost / Capital Cost	1	0.1

4. Technology of CO_2 Reuse in the Production of PCC

CO_2 as an industrial gas can be used (or reused) in many applications in different technologies. One of the direct chemical applications is the production of precipitated calcium carbonate (PCC), which we are presenting here. PCC represents a high-value product with a competitive local market.

PCC technology demands serious research on interfacial phenomena[10] and rigorous scale-up techniques from the laboratory, to the pilot scale, and then to industrial production.[10] Several questions appear when research involving pure chemicals on the laboratory scale is performed and when the principles are transferred to the industrial scale, based on real raw materials. The demands of the product relating to different parameters, like size distribution, crystal structure, zeta-potential, rheology, porosity, retention time, and whiteness, are very rigorous and need a serious approach in order to be fulfilled. As a consequence, some very rigorous product properties, equipment specifications and process conditions, are demanded.

The engineering modeling of product quality and price, as well as the modeling of production time, dependent on relevant properties and conditions, is of importance for scale-up procedures.

The PCC process has been realized as an extended pilot-plant unit. After the initial pilot testing phase, the

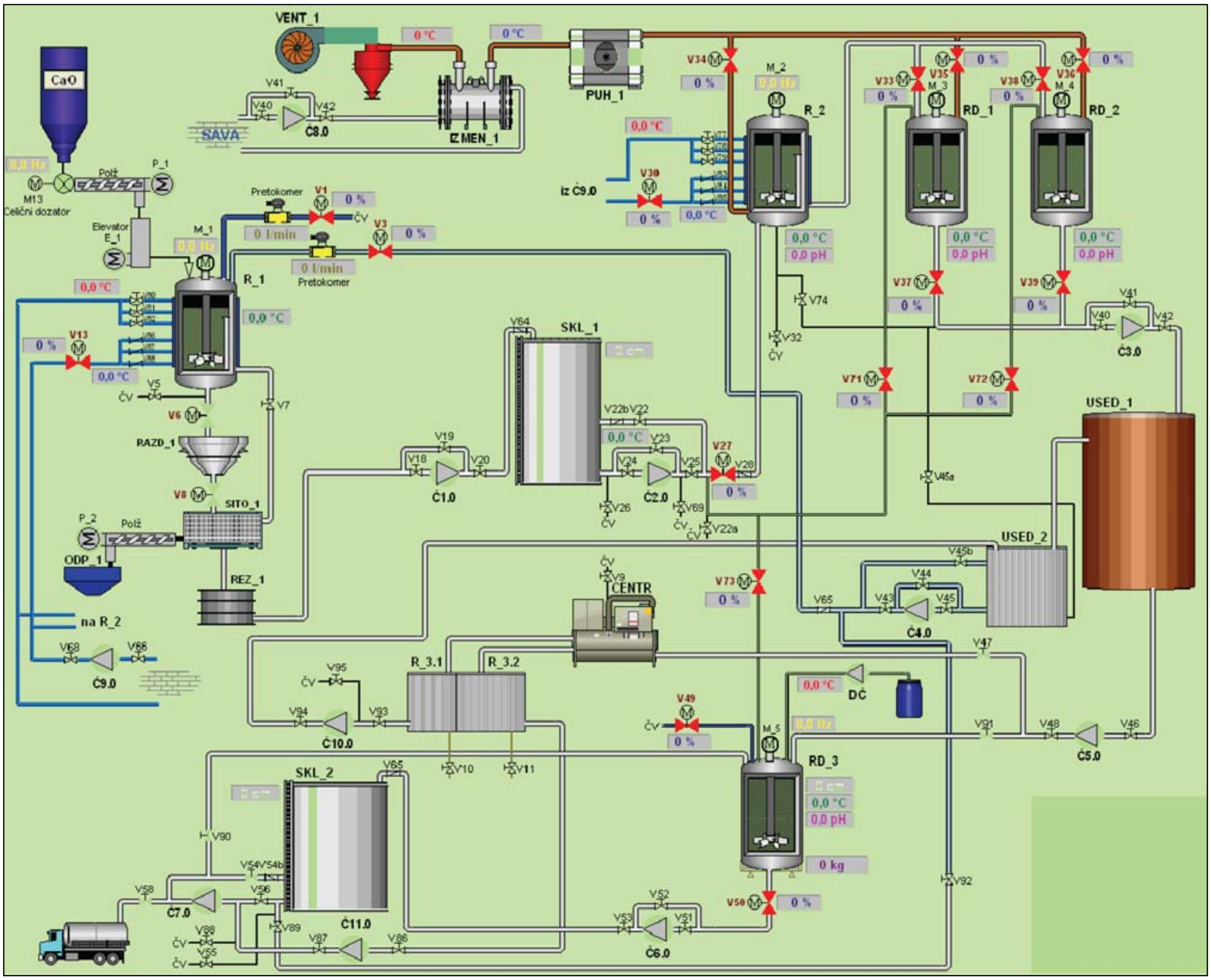

Figure 3: Screen copy of the SCADA page of the Industrial Control System for the PCC process

process was upgraded and arranged to become a full operating process, presentation of real process is done by screen copy of our SCADA program page – Figure 3.

5. Fly Ash as CO_2-Capturing Media

Fly ash, as an end product of the coal-burning process, comes from thermal power plants in large quantities.[12] Some amounts of fly ash are used for different industrial applications[12–14] like cement, but most of them are deposited.[14] From the results of a chemical analysis of typical fly ash from different origins, presented in Table 2, one can find alkaline earth metal oxides as a possible potential for reaction with CO_2, what is in most cases not utilized. Polish researchers have been very active[15] in the past, trying to examine and apply the potential of fly ash for CO_2 capturing.[13]

The theoretical potential of fly ash, of Slovenian origin, for reaction with CO_2 has been calculated from the basic reactions (3) and (4) and data from Table 2. Theoretical capacity of fly ash for CO_2 capturing is 170 kg/t. In this way, a mixture of fly ash and gypsum, which is deposited in mine tunnels, may be upgraded by the conversion of the alkalinity in fly ash to carbonates. As Slovenian thermal power plants produce of about 1.1 Mt/a of fly ash[12] such possibility has a significant potential. Calculations show possibility to sequester of about 131 kt/a of CO_2 at 70% efficiency.

The mineral carbonation reaction system of fly-ash components with CO_2 is relatively complex. In the first stage, during the hydration phase, the $Ca(OH)_2$ – portlandite and C–H–S phases form, which finally react with CO_2:

$$Ca(OH)_2 + CO_2 \rightarrow CaCO_3\downarrow + H_2O \qquad (3)$$

$$CaOnSiO_2mH_2O\ (C\text{–}S\text{–}H) + CO_2 \rightarrow CaCO_3\downarrow + nSiO_2 + mH_2O \qquad (4)$$

Besides calcium compounds, other oxides, like MgO, contribute to CO_2 absorption in fly ash.

To realize this idea, a research-development-technology market approach, typical for a product-engineering procedure, is necessary.

Table 3: Chemical composition of Slovenian* and some Polish fly ash[13,15]

Ash sample origin:	Slovenia*	Poland -FAFB[13]
Component:	*w*/%	*w*/%
SiO_2	47.08	34.2
Fe_2O_3	10.01	7.4
Al_2O_3	19.36	16.2
CaO	14.29	10.5
MgO	2.85	3.74
SO_3	1.15	6.49
Na_2O	1.28	0.75
K_2O	1.91	1.42

* Source: industrial analysis report

6. Algae Growth

A rapid expansion in the global technological activities associated with CO_2 consumption for algae growth has been recorded in the past several years. Among the numerous studies on algae growth, the U.S. DOE study[18] from 2010 covers the widest range of problems in the sense of presenting engineering problems, application possibilities and the state of the art.

Algae production does not actually sequester fossil carbon, but rather it provides CO_2 capture and its reuse in the form of fuels and other products that are derived from the algae biomass.

Researchers have been working to create viable processes to derive biofuels from algae. Several pilot farms are operating around the world, and many more are planned; however, most of them are small-scale demonstration projects. The basic questions about algae growth rate and lipid yield and its effective separation from the algae are still being very intensively researched on the laboratory and pilot scales.[19–21] However, among the various technological solutions, we can find two typical cases of successful pilot engineering applications.[22,23]

For studies on algae science and engineering applications, an interdisciplinary approach based on biology and engineering is necessary. Our work takes into account the global research programs related to algae systems across a broad spectrum, from scientific topics to technological solutions. We have designed a small pilot unit to study the process conditions for algae, the production capacity and the content of oil. Future advanced studies on algae, its lipid yield, content and composition, are challenges relating to the production capacity.[23,24]

6. 1. The Algae Growth Program at FKKT Ljubljana

Besides the use of CO_2 in the process of photosynthesis, which actually means the sinking of CO_2 into new cell constituents, algae are organisms that do not necessarily need agricultural land for the development of their biomass. As we see in nature, they can survive in different ecosystems, such as thermal springs or on the surface of snow, using various nutrients that can be found in fresh and sea waters or on soil surfaces. If one provides special cultivation conditions and concentrations of available nutrients, algae could produce large amounts of proteins, carbohydrates and lipids. The recovered algal biomass can be easily processed into many products, such as animal feed, fertilizers, chemicals, food, medicines, or converted to biofuels, such as bioethanol and biodiesel, or simply used for biogas digestion.[22]

Algae are eukaryotic macroalgae and microalgae, but also prokaryotic autotrophic specie, nowadays referred to as cyanobacteria. The advantage of algae, especially microalgae, lies in their metabolic flexibility, with possible modifications to their metabolic pathways and cellular contents. By changing the parameters that influence growth, the cell biosynthesis turns towards the augmentation of proteins, carbohydrates or lipids.[25]

The laboratory biomass production of algae was studied with Chlorella vulgaris SAG 211-12, obtained from the algal collection of the University of Göttingen. The algal production was studied in batch cultures (3 l capacity), placed on a laboratory vessel with a stirrer, a 2600 Nalgene Culture Vessel with ports (15 l capacity) and a custom-made laboratory pilot plant, composed of a vessel with stirrer (capacity 50 l) and a coil of plastic tube positioned around a support for fluorescent PAR lamps (capacity of 9.4 l). In the first and second cases the culturing vessels were placed in a thermostat-regulated at room temperature (22–24 °C) with 12 h/12 h day and night period. The laboratory pilot vessel was maintained at room temperature. In the second and third cases the pH value (7.0) was controlled by CO_2 gas injection.

The main objective of the study was to establish the parameters of growth for enhanced lipid production, which was reported for Chlorella vulgaris in the range w = 14–40% (even 56%).[26] At the beginning we propose an enriched culture medium (Jaworski algal medium) based on a modified Solvay solution of hydrogen carbonate (γ($NaHCO_3$) =1.16 g/l) what represent the sink for the CO_2 gas.[27] Bat in batch cultures with a proposed medium, the algal concentration was achieved in the range between 7.1 $\times 10^6$ and 2.1 $\times 10^7$ cells in a ml of culture for an photon flux density (PFD) of 80 μmol/s m^2 to 120 μmol/s m^2, and only w = 1.69% of lipids were determined in the dry biomass due to the unbroken cell walls of the chlorella cells.

In proceeding experiments the Nalgene Culture Vessel 2600 was used for culturing the chlorella strain.

The vessel contained 15 l of low-nitrogen growth medium ($\gamma((NH_4)_2HPO_4)$ = 20.3 mg/l, $\gamma(KCl)$ = 223.6 mg/l, $\gamma(MgSO_4\ 7H_2O)$ = 504.5 mg/l, $\gamma(KH_2PO_4)$ = 136.1 mg/l and $\gamma(FeSO_4 \cdot 7H_2O)$ = 1.83 mg/l). Experiment started with 2.16×10^6 algal cells/ml. The culture was aerated at an airflow rate of 270 l/h and a pH value of 7.0 was maintained by the injection of CO_2 at a flow rate of 2 l/h. Medium was mixed with a Rushton turbine at 2.5 Hz. The pH, dissolved O_2, redox and *temperature* values were registered during the experiment. The photon flux density (PFD) during the day period was in the range from 50 μmol/s m^2 to 183 μmol/s m^2, depending on the quadrant of illumination. The experiment was finished after 200 hours.

In the first six experiments the influence of the day and night periods on the production of lipids was studied. The day (h)/night (h) periods (ratio) were chosen as follows: 16 h/8 h, 20 h/4 h, 24 h/0 h, 12 h/12 h and 8 h/16 h.

The maximum yield of algal concentration was achieved for the illumination regime 16 h/8 h, with a final 4.5×10^7 cells/ml, but the maximum yield of chlorophyll was for the illumination regime 8 h/16 h, with 1.6 mg/l. The production of lipids was the highest in the case of the 16 h/8 h illumination, with w = 27% of dry weight of algal biomass, and the lowest for the illumination regime 20 h/4 h, with w = 19% of dry weight of algal biomass.

The variation of nitrogen nutrients, nitrate source and ammonia source, was studied in 13 two-week experiments to establish the best combination of limited nitrogen source for lipid production, while the other components of the culturing medium were from the Jaworski medium. The values of $\gamma(NaNO_3)$ were in the range from 8 mg/l to 800 mg/l, and for $\gamma((NH_4)_2HPO_4)$ were in the range from 2.03 mg/l to 203 mg/l. The highest yield of total lipids (w = 36.7% of algal dry weight) was produced with a combination of 124 mg/l of $\gamma(NaNO_3)$ and 31.5 mg/l of $\gamma((NH_4)_2HPO_4)$, but the basic low-nitrogen medium with 8 mg/l and 20.3 mg/l, respectively, yielded the maximum amount of total lipids fraction in algae of w_{max} = 38.2%.

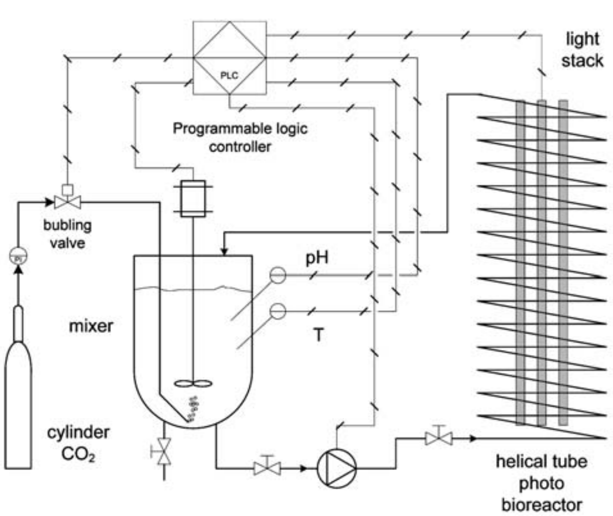

Figure 4: Diagram of pilot spiral bioreactor for algae production

In the laboratory pilot plant – Figure 4 only one experiment was concluded with the medium composition: $\gamma(NaNO_3)$ = 58.7 mg/l, $\gamma((NH_4)_2HPO_4)$ = 31.461 mg/l and the rest of the compounds in the same concentration as in the Jaworski medium, which yielded the maximum algal concentration in the previous experiments. The logarithmic phase of growth was maintained with a withdrawal of a part of the algal biomass on the fourth, seventh and ninth days of the pilot plant's operation and the replacement with a fresh medium. The experiment was stopped on the fourteenth day because of the outgrowing of the plastic tube coil with attached algae. Nevertheless, the maximum specific growth rate was maintained in the range of 0.58 d^{-1} to 0.69 d^{-1}.

7. Conclusion

In this contribution we wanted to emphasize the concept of a product-engineering approach to the effective transfer of research results from laboratory experimental work, to a pilot plant's development, and then to an industrial application. Different methods of CO_2 capturing, like inorganic systems based on the activation of calcium ions and biological systems based on algae, are promising challenges for engineers working closely with chemists and biologists.

8. Acknowledgment

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Povzetek

Človeštvo je v zadnjih 200 letih povzročilo enormno rast koncentracije CO_2 v ozračju. Mednarodna skupnost je pričela z ukrepi za zmanjševanje emisij CO_2, zato morajo posamezne države sprejemati vse več obveznosti glede tega. V članku opozarjamo, da zmanjševanje emisij CO_2 lahko za nacionalne ekonomije predstavlja tudi novo priložnost za pospeševanje lastnega razvoja in ne le grožnjo. Analiziramo slovenske razmere, ki ponujajo priložnosti za pospeševanje trajnostnega razvoja na podlagi lokalnih virov ter hkrati omogočajo usmeritev v nizkoogljično družbo. Tehnologij, ki so že prisotne v Sloveniji, kot je koncentriranje CO_2 z aminskimi procesi še ni mogoče ekonomsko učinkovito prenesti na bistveno večje sisteme kot so termoelektrarne, zato je nuja po postopnih korakih toliko večja. Ena od možnosti predstavlja uporaba CO_2 v industrijskih procesih kot na primer za izdelavo oborjenega kalcijevega karbonata. Drugo možnost prinašajo alkalne komponente v elektrofiltrskem pepelu iz termoelektrarn, ki lahko vežejo znatne količine CO_2 in se sedaj v nevezani obliki odlagajo v rudnike. Pomembne priložnosti odpirajo biotehnologije od samega gozdarstva do gojenja alg. Z načrtnim pogozdovanjem (ne zgolj zaraščanjem) je mogoče zajeti pomembne dodatne količine CO_2. Lesna in druga biomasa, tudi alge, lahko v bodoče predstavljajo pomemben lokalni vir in tudi zamenjavo za fosilna goriva, predvsem pri ogrevanju, kar bi pomenilo dodatno zmanjševanje emisij CO_2. Za učinkovito pretvorbo tehnoloških priložnosti v poslovne priložnosti je potreben proces inženirizacije zamisli – prenosa znanja iz laboratorijskega merila preko pilotnih naprav v veliko industrijsko merilo. Sodelovanje tako naravoslovnih kakor tehniških raziskovalcev (inženirjev) je tako nujno potrebno