

Bioelectrosynthesis as an Alternative to Photosynthesis

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Received April 26, 1991; Accepted January 28, 1992

ABSTRACT

The CO₂ reduction processes have been discussed as a way of designing an ecologically totally closed technology. An electric current and molecular hydrogen are the two related available agents that can be discussed as ecologically pure reductants. The most important products are liquid and gaseous fuels, the products of large-scale organic synthesis, monomers, and amino acids. For CO₂ reduction, the necessary energy consumption and H₂ costs were calculated. For complex organic molecules, amino acids for instance, the energy consumption does not make up the main portion of the costs.

The biocatalytic systems of CO₂ reduction based on cryoimmobilized cells are described. Conversion of CO₂ into L-lysine with electrochemical decomposition of water was effected on the laboratory scale. A general unit for diverse technological processes can be a bioelectrosynthetic modulus, an electrochemical hydrogen generator coupled with a biocatalytic converter of hydrogen and oxygen. The systems for bioelectrosynthesis of motor fuels and essential amino acids have been economically estimated and characterized. The possibilities of combining the solar energy transformation and H₂-CO₂ conversion have been discussed.

Index Entries: Bioelectrosynthesis; CO₂ reduction; liquid fuels; amino acids; immobilized cells; economic estimates.

INTRODUCTION

Recent decades clearly have shown some hazards brought about by modern industry. The ever-intensifying production of energy, the evaluation of CO₂ discharge into the atmosphere, and an appreciable depletion of easy-to-mine mineral fuels are well known negative global tendencies. Naturally, the development of energetics should not provide an infinite growth of energy production but must be associated with renewable sources and increasing the efficiency of the energy conversion processes. Humankind has learned to convert efficiently thermal energy into mechanical energy and electric, atomic, or thermal energy into electricity. Currently, intensive work is being carried out on designing systems for solar energy conversion.

A global problem is that recent decades have faced increasing energy consumption in the production of food. Energy consumption has grown because of the application of fertilizers, mechanization, food processing, and distribution (1-3). Thus, the present-day cost of a food calorie is 10 "simple" heat calories. A tendency to its growth aggravates the situation (4-7). This influences the cost of various forms of energy. If the simple forms of energy are fairly cheap (for instance, approx 1¢/kW·h of electric energy), the energy of food products costs 50¢-\$5 per kW·h.

On the other hand, a fundamental problem of global ecology seems to be an accumulation of carbon dioxide as a product of the combustion of mineral fuels. Current energy-producing technologies keep increasing CO₂ discharge into the atmosphere. The consumption of mineral fuels yields about 5·10⁹ tons carbon annually in the form of CO₂ into the air. The aftereffects of this process are well known and have been discussed in the literature (4-7). The observed (4-7) ominous elevation of global temperature is one of the calamitous environmental shifts.

The observed nonstop increase of CO₂ in the atmosphere shows that photosynthesis cannot cope with CO₂ evolution processes (7). There are no industrial technologies now for CO₂ conversion into useful products. Designing such technologies is one of the most urgent and promising tasks of science and technology.

What are the most interesting approaches in this respect? What economy restraints should be regarded when designing a large-scale industrial technology for CO₂ reduction? What are the benefits of feasible technologies compared to natural photosynthesis? What are applicable technologies for CO₂ reduction? What is the role of the biocatalytic approach in solving the problem of CO₂ conversion? The objective of this work is to consider the possibilities of bioelectrosynthesis as a device for CO₂ reduction. In a larger sense, this is an effectuation of biocatalytic processes. Their driving force providing the thermodynamics of the reaction is the electric energy. Potentially, bioelectrosynthesis can be realized as both a direct electrode reaction with enzymes and an intermediate electrolytic production of hydrogen. The second path seems to be notably simpler.

THE CONCEPT OF HYDROGEN-CO₂ ECONOMY

The chemical conversion of CO₂ into various products can be effected by various reducing agents. If the problem is in the designing of the technologies capable of increasing CO₂ consumption, the conversion processes should be large-scale and yield the compounds broadly marketed. The important conditions should be the availability and extreme ecological purity of a reductant. Analysis shows that there are only two related agents meeting these demands: electric current and molecular hydrogen. The concept of "hydrogen economy" is fairly well scrutinized in the literature. The advantages of hydrogen as an available, energy-intensive, ecologically pure, and transportable energy source are discussed in detail in many papers, reviews, and monographs (8-16). The analysis given below shows that a large-scale application of hydrogen can be economically valid even now, if hydrogen can be applied as CO₂ reductant.

The cheapest and most contemporary method of hydrogen production is the electrolytic decomposition of water (15). Ecologically, electroreduction of CO₂ is attractive because it uses energy unrelated to burning an organic fuel. These can be nuclear or solar energies. The methods of hydrogen production by coal-steam conversion do not seem to be ecologically fair and are economically inferior to the electrolysis of water.

For the most attractive reactions of CO₂ reduction, we calculated the necessary energy consumption and H₂ costs for the process (Table 1). The most important products were selected. These are: liquid and gaseous fuels, products of large-scale organic synthesis, monomers as the basis for production of plastics and rubber, and food products.

Theoretical energy consumption is given in kW·h/kg of product and corresponds to the electric energy of direct CO₂ reduction at the potential of equilibrium hydrogen electrode. The cost of electrolytic hydrogen (15), is assumed to be 2.52¢/kW·h and that of photohydrogen obtained by photovoltaic decomposition of water 9¢/kW·h. However, estimates can vary; so, in some places in Northwest US, the cost of hydropower might be as low as 3¢/kW·h. The variations can change the particular economic figures but cannot alter the general approach.

It can be seen in Table 1 that the reduction of CO₂ is very energy-consuming. However, this assumption is, in most instances, offset by the cost of the final product. In this case, the more complex the organic molecule, the more economical the production of the substance. Thus, the production of 1 kg L-lysine theoretically calls for 6.29 kW·h of electric energy. If one takes into account that the cost of lysine (commercially produced at present as a fodder additive by microbiological conversion of carbohydrates) is \$3-10 per kg, it becomes evident that energy consumption does not make up the main portion of the costs. The most essential problem becomes the creation of efficient and selective catalysts of CO₂ reduction.

Table 1
Reduction of CO₂ into Various Products:
Energetics and Economic Estimates

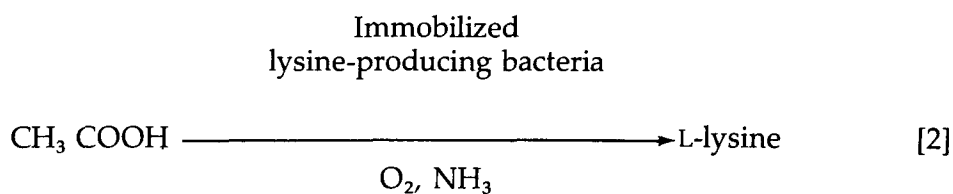
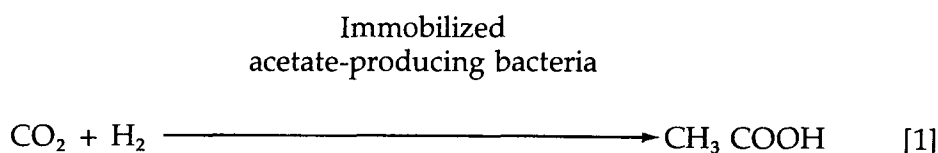
Product	Theoretical consumption of energy (kW·h/kg)	Cost of electrolytic hydrogen per unit of product (cent/kg)	Cost of photohydrogen per unit of product (cent/kg)
Hydrocarbons			
Methane	16.40	41.32	147.00
Ethane	15.31	38.60	137.00
Propane	14.91	37.60	134.00
Hexane	14.49	36.50	130.00
Octane (isooctane)	14.38	36.20	129.00
Monomers			
Ethylene	14.06	35.40	126.00
Propene	14.06	35.40	126.00
Butene	14.06	35.40	126.00
Butadiene	13.36	33.70	120.00
Isoprene	13.50	34.00	121.00
Alcohols and Glycols			
Methanol	6.15	15.50	55.40
Ethanol	8.56	21.60	77.00
Propanol	9.84	24.80	88.60
Butanol	10.51	26.50	94.60
Ethylene glycol	6.56	16.50	59.00
Glycerol	4.99	12.60	44.90
Organic acids			
Formic acid	1.42	3.57	12.78
Acetic acid	4.37	11.00	39.30
Propionic acid	6.62	15.60	55.80
Butyric acid	7.45	18.80	67.80
Ketones, Esters			
Acetone	10.18	25.70	91.60
Ethyl acetate	8.94	25.50	80.50
Carbohydrates			
Glucose	4.37	11.00	39.30
Amino acids			
Methyonine	2.43	6.10	21.90
Aspartic acid	2.96	7.50	26.60
Histidine	4.23	10.70	38.10
Glycine	4.37	11.00	39.30
Threonine	4.41	11.10	39.70
Alanine	4.42	11.10	39.80
Lysine	6.29	15.90	56.60
Tyrosine	6.88	17.30	61.90
Tryptophan	7.69	19.40	69.20
Phenylalanine	7.89	19.80	70.80

Prices of hydrogen are taken from Ref. 15 (2.52 ¢/kW·h).

BIOCATALYTIC SYSTEMS OF CO₂ REDUCTION

Heterogeneous Catalysts on the Basis of Cryoimmobilized Cells

Analysis shows that the most beneficial products are amino acids (Table 1). This makes evident the following: The search for ways to reduce CO₂ emissions should be related to the creation of catalysts on the basis of enzymes and enzymic systems. We have found that this problem is solved by using the immobilized microbial cells, superproducers of necessary products. Consider the features of catalytic processes of this type by the example of the catalytic system of reduction of CO₂ into L-lysine. The effectuated scheme of the process is:



At the first step, the immobilized anaerobic acetate-producing bacteria act as a catalyst. The second step is the conversion of acetate into L-lysine by immobilized *Corynebacteria* strains, lysine producers. We used the method of cell entrapment in a polymer matrix via cryoformation of a polymeric gel (17). The method consists of freezing the cell suspension in a solution of polyvinyl alcohol to -20 – -30°C with subsequent temperature elevation and activation of the biocatalytic systems. This procedure results in mechanically strong and highly permeable gels with embedded cells. The method has been tested on multiple systems, including the system of ethanol production on the basis of the immobilized bacterial and yeast cells as well as systems for the production of cellulases, hydrogen, acetic acid, amino acids, and some vitamins (17). The method is also applicable for immobilization of animal and human cells.

The elaborated heterogenous catalytic systems have some principal advantages.

1. The resultant heterogenous catalysts feature a prolonged work without a notable decrease in the catalytic activity. Thus, we used *Acetogenium kivui* culture for 2 yr with 80–90% initial activity retained. Cryoimmobilization of cells affords the work in semisterile and partially aerobic conditions.

2. Anaerobic producers of acetate are slowly growing cultures. At low growth parameters, the processes on free-growing cultures will be extremely unproductive. The limitation of the processes based on free-cultivated cells drastically lowers the productivity, and in the case of slowly growing cultures, the productivity is insignificant. The immobilization of cells obviates this limitation (18–20).
3. The material (polyvinyl alcohol) used for immobilization is an inexpensive and available polymer. This permits large-scale implementation of the process.
4. The developed method of cell immobilization is a unique procedure for immobilization of thermophilic microorganisms. The temperature range of the catalytic activity is 0–70°C. The known methods for cell entrapment in gels using carrageenin or alginate afford stable structures up to 40–45°C (21–23).
5. The immobilized system features a high capacity for recovery. After the catalyst has been worked out and inactivated, the cryogel can be melted at 90–100°C, and the polymer can be used for a repeated immobilization.
6. Cryoimmobilization affords the catalysts containing up to 10% cell biomass. So, the activities of the catalysts are fairly high. Theoretically, the process can be organized in the form of a complete utilization of hydrogen. The immobilized cells either do not grow at all or grow very slowly. So, there is no loss of efficiency at the step of catalytic conversion.
7. On the basis of cryoimmobilization, various structures can be formed, such as spheres, films, layers, and polylayers, the particles of regular and irregular shape.

The above example demonstrates a principal possibility for creating high-efficiency catalytic systems capable of reducing CO₂ to complicated molecules, such as amino acids. By using the methods of microbiology, selection, molecular genetics, and genetic engineering, catalysts can be created for most of the reactions listed in Table 1.

Further development should be related to the expansion of catalytic forms and reactions as well as to the designing of the high-efficiency reactors of the process.

BIOELECTROSYNTHETIC MODULUS

Technical solutions and construction of reactors for conversion of CO₂ to desired products can vary. As production unit, there can be:

1. A plant for production of fuel, organic synthesis products, or amino acids as food;

2. A modulus at a cattle breeding farm for a flow conversion of carbonic acid into essential amino acids;
3. An isolated solar converter in a desert producing fuel or food; and
4. The closed systems for vital activity, to convert an electric, solar, or fuel energy into energy-rich products and oxygen.

A general unit, for all these diverse technological processes, can be a bioelectrosynthetic modulus, an electrolytic device coupled with a biocatalytic converter of hydrogen and oxygen.

As basis, consider an industrial electrolyzer, a hydrogen and oxygen generator, and the parameters of the processes for production of liquid fuel and amino acids. The parameters of an industrial EL-250 generator are the following: the power, 185 kW; the productivity, 42 m³ H₂ and 21 m³ O₂/h; and the energy efficiency, more than 80% (15). Generators of this type feature high reliability, simple control, full automation, and over 20 yr in operation.

Bioelectrosynthesis of Motor Fuel

Hydrogen productivity of the system 42 m³ H₂/h corresponds to 45 · 10³ mol hydrogen a day. Upon conversion of this amount of hydrogen via CO₂ reduction, one can obtain 480 kg methanol, 345 kg ethanol, 330 kg ethylacetate, or 205.2 kg isooctane per day. At the price of motor fuel (40¢/L), an electrosynthesizer of this kind can yield a daily product costing \$100–200, for which it consumes 4.5 · 10³ kW · h of electric energy. The cost of the electric energy consumed is \$50/d. It is possible to imagine a petrol station working on the electric energy and having 20 fuel electrosynthesizers. This petrol station could produce about 8000 L motor fuel per day. Its realization could bring at least \$1 · 10⁶/yr at the price of fuel, 40¢/L. The relevant electric energy costs make up \$360,000/yr.

To date, it is difficult to estimate the cost of creating an infrastructure for motor fuel production using this technology. However, the bioelectrosynthesis of motor fuel has two strategic advantages: The technology affords, in principle, a steady economy independent of outer sources of mineral liquid raw materials; and the exhaustion of world oil sources causes the price for petrol raw material to rise, which makes the electrosynthesis of liquid fuel strategically and economically valid.

Bioelectrosynthesis of Essential Amino Acids

Consider the possibilities of a bioelectrosynthesizing modulus in the production of some essential amino acids finding a vast and fast-growing market as fodder for animals. Many amino acids are used as food additives for humans as well as in the production of some drugs and medical devices.

The output of a bioelectrosynthesizer, equal to 42 m³ H₂/hr, should theoretically provide a daily output of 469.3 kg L-lysine, 383 kg L-tryptophan, or 1215 kg L-methionine. Current prices for fodder lysine are \$3–5/kg and for fodder tryptophan or methionine, \$8–10/kg. Thus, only one bioelectrosynthetic modulus using the EL-250 hydrogen generator can provide the minimal production of lysine, tryptophan, and methionine (annual costs about \$0.5, 1.1, and 3 million, respectively). The costs of the electric energy needed for CO₂ reduction make up an incommensurately small value, about \$20,000/yr.

The bioelectrosynthetic modulus can be installed directly in an agricultural farm to transform the CO₂ to a solution of essential amino acids that can be used as fodder additives without isolation and additional purification. This makes the cost of the product extremely low. Thus, the bioelectrosynthesis of the compounds being energy-rich metabolites is economically valid and potentially highly profitable.

SOLAR ENERGY

Hydrogen–CO₂ Conversion

Table 1 lists the costs of hydrogen derived by photoelectric conversion and spent on CO₂ reduction to synthesize various products. It shows that the bioelectrosynthesis of fuels (methanol, ethanol, and octane) should cost 2–3 times more than present-day prices for fuel. Nowadays, no one dares to prognosticate the dynamics of oil prices. It is quite possible that in the future, the solar energy–fuel conversion will prove economically beneficial.

An utterly different situation occurs when solar energetics is coupled with the synthesis of energy-rich metabolites. Table 1 shows that when using photovoltaic hydrogen, the energy cost in the synthesis of amino acids is 20–70¢/kg product. At the product price of \$2–10/kg, amino acid production using solar energy becomes economically profitable. Such solar energy hydrogen–CO₂ converters installed in semidesert regions can provide quite an intensive agricultural production.

BIOELECTROCATALYSIS AND DIRECT CO₂ REDUCTION

The above-discussed schemes of CO₂ reduction are based on the idea of using conventional technologies for production of hydrogen as a universal reductant. The overall scheme of the process is: the production of hydrogen and oxygen in an electrolytic cell—utilization of hydrogen and oxygen in separate reactors. Of interest is the elaboration and implementation of devices to directly reduce CO₂ in an electrochemical cell.

Biocatalytic systems as catalysts of electron transfer at the interface electron/ionic conductor have long been of interest to our group of researchers (24–33). We reported the first successful direct electrochemical electron exchange between electron conductors and enzyme active sites (23). On the basis of immobilized enzymes, we created the electrodes for electroreduction of oxygen and electrooxidation of molecular hydrogen. Earlier publications (33,34) reported the prospects for designing the bioelectrocatalysts working in the electrosynthetic system, i.e., transforming the electric energy into the energy of chemical bonds.

Bioelectrosynthesis with direct electrode CO_2 reduction can have certain advantages. Well organized, the process can be run at the equilibrium potential, and thus, energy consumption can be brought to theoretical values (Table 1). A second important feature of bioelectrosynthesis with a direct CO_2 reduction is the possibility of running the electrosynthesis directly in the medium of various electrolytes, for instance in sea water or human or animal blood. In the latter instance, the bioelectrosynthesis can be a basis for electrofeeding animals or humans. Bioelectrosynthesis can reverse the process of carbohydrate oxidation and convert exhaled carbon dioxide to carbohydrates (or related products) with oxygen saturation of blood. The project looks quite fantastic. Nevertheless, designing systems of electrofeeding living things seems feasible. The power of a human is 100–200 W. Hence, a direct electrofeeding of a human population (about $6 \cdot 10^9$ persons) needs an electric power station of $6 \cdot 10^9$ MW.

CONCLUSIONS

Agricultural production, as a method of energy conversion, uses thermal, solar, and electric energies to produce energy-rich metabolites. The costs of initial forms of energy are rated in $\text{¢/kW} \cdot \text{h}$. The costs of agricultural products are rated in $\text{\$/kW} \cdot \text{h}$. Agricultural production from the viewpoint of an energy supply of humans and animals has no alternative. Bioelectrosynthesis can be a potential alternative.

Bioelectrocatalysis, performing the synthesis of energy-rich metabolites (carbohydrates and amino acids), provides a unique possibility for production of food and fodders by direct conversion of electric energy. The conversion efficiency depends on electrolytic decomposition of water and can be 70% or more. If only 10% energy, spent currently in agricultural production, can be used via bioelectrosynthesis having 60–70% efficiency, it is possible to derive the products energetically more valuable than the same amount produced by contemporary agriculture.

It is important to emphasize once again a principal feature of bioelectrosynthetic systems: Carbonic acid is reduced with the formation of molecular oxygen. Chemically, the above processes correspond to an overall reaction of photosynthesis. However, they proceed not under the action of solar energy, characterized by low flow densities, but under that

of electric energy. Hopefully, this opens the way to designing high-intensity industrial processes. Realization of these processes depends on a new level of insight into the catalytic reactions and on moving toward the biocatalytic systems capable of providing a high efficiency in deriving complicated molecules.

In conclusion, it should be pointed out that the idea of designing systems for bioelectrocatalytic CO₂ reduction is at the start of its development. This idea seems very tempting. It is unique and central in solving the problem of designing an ecologically totally closed technology. It provides a way for conversion of a "simple" energy to energy-rich metabolites as the ground for high-efficiency production of foods. It follows from this paper that this idea should be highly attractive from an economical viewpoint.

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Errata

The following are the corrected figure captions for Ramos, L. P., Breuil, C., and Saddler, J. N. (1992), "Comparison of Steam Pretreatment of Eucalyptus, Aspen, and Spruce Wood Chips and Their Enzymatic Hydrolysis," *Appl. Biochem. Biotechnol.* **34/35**, 37-47.

Fig. 2. Enzymatic hydrolysis profiles of steam-treated substrates derived from eucalyptus wood chips. Hydrolyses were carried out at (A) 2% and (B) 10% (w/v) substrate concentrations using 10 FPU g⁻¹ cellulose. (□) SEE-WIA, alkaline-insoluble fraction; (▲) SEE-WIA/H₂O₂, peroxide-treated fraction.

Fig. 3. Comparison of the hydrolysis profile of the peroxide-treated fractions derived from steam-treated (▲) *E. viminalis* (SEE-WIA/H₂O₂), (□) aspen (SEA-WIA/H₂O₂), and (■) spruce (SES-WIA/H₂O₂) wood chips. Hydrolyses were carried out at (A) 2% and (B) 10% (w/v) substrate concentrations using 10 FPU g⁻¹ cellulose.