# Formate Cycle: The Third Way in Green Energy

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***Abstract*****—The prospects of the formate cycle are considered: the fixation of $CO_2$ from the atmosphere in the form of formic acid in low-temperature electrochemical reduction, and further use of formic acid (formates). A brief review of effective catalysts (in particular - without the use of platinoids) for the direct oxidation of formates in a fuel cell and for the decomposition of formates with hydrogen evolution is given in the aspect of "green energy". Perspectives of formates, as a hydrogen accumulator, and as a carbon monoxide (CO) accumulator are shown. Particular attention is paid to the dual use of formate, as a combined raw material for the Fischer-Tropsch process, which allows a simple, technological and cost-effective production of synthetic liquid fuel (ergic-gasoline) suitable for classic automotive engines. It was suggested that formic acid should be used as an energy carrier produced in solar or wind power plants as a buffer reserve or a commercial product.**

***Index Terms*****—Formic acid, hydrogen accumulator, Fischer-Tropsch process.**

## I. INTRODUCTION

In work [1] the general prospects of the cycle of $CO_2$ fixation from the atmosphere in the form of formic acid or formates in a low-temperature electrochemical process, which is reasonably economically rational, were considered. Formic acid can serve as a safe renewable hydrogen source (hydrogen battery), or used in a fuel cell [2]. Also, the use of formic acid as a renewable raw material for environmentally friendly production in organic chemistry and synthetic liquid fuel for classic internal combustion engines was considered.

Separately, the process of carboxylation was discussed, in which each mole of formic acid additionally binds a mole of $CO_2$ to produce oxalic acid (also a potential raw material of combating greenhouse gases. The alarmist sentiment in organic chemistry). This process is interesting in terms of burdensome ways of such a struggle society leads to the promotion of economically.

The same can be said about environmental alarmism in general, and economically onerous ways of "green energy". Formation energy, as will be shown below, has economic prospects, so its development does not depend on the mode of alarmism. This article systematizes the theses about what preconditions are necessary for the wide use of the formate cycle, and which of them already exist.

## II. CHEMICAL AND TECHNOLOGICAL PRECONDITIONS FOR THE FORMATE CYCLE

In any process of chemical (or electrochemical) synthesis, developed for mass application, the question of the catalyst is critically important. This applies both to the technology for the production of the catalyst, and to the process itself, in which the catalyst is used. The catalyst should:

1) not require super-expensive or obviously rare raw materials for production.

2) not require supercomplex and super-expensive technology for production.

3) not require exotic working conditions for temperature and pressure.

4) not create a high risk in production and especially in the working operation process.

5) have sufficient resistance to the influence of changes in the state of the environment

6) Have sufficient durability under normal operating conditions

In the field of application of formic acid as an energy carrier for fuel cells (DFAFC or DFFC - direct formic or acid fuel cells), as for many other processes of anodic electrochemical oxidation of hydrogen and organic energy carriers, the compounds of platinum or platinoids have traditionally been considered as catalysts. Before talking about a change in the situation in this area, we should dwell on the "platinum problem" for all serial fuel cells on hydrogen and organic energy carriers.

Platinum metals are very rare and expensive, and this generates tough price barriers. In the case of the use of fuel cells in miniature devices (such as mobile phones and microcomputers), the price of the catalyst does not really matter - it is in any case very small part of the total price of the device. In the case of larger devices - for example, cars, the price of such a catalyst becomes extremely significant. The cost factor of catalysts for the automotive industry was clearly manifested in dieselgate - although there it was a catalyst only for the secondary oxidation process (afterburning of CO after the internal combustion engine). In the case of cars (more precisely electric vehicles) on fuel cells, the catalyst is necessary for the primary oxidation process, i.e. for, in fact, energy production, and there the catalyst factor may prove to be one of the determining factors for the price (and hence for the competitiveness) of the product. Even more significant is the price of the catalyst for ship power plants using fuel cells (like the German U212A submarine), or for local fuel cell power plants (already limited in the market today).

Shortly about this submarine. The total length of the 212A

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submarines in the basic version of the project is 56.08 m, width - 7 m, normal draft - 6 m. In the above-water position, the displacement is 1580 tons, in the submerged - 1990 tons. More than 10 submarines are already in operation

For secret motion at depth, the Type 212A submarines can use their power plants in an air-independent mode. In this case, 9 oxygen-hydrogen fuel cells based on polymer electrolyte are used as an energy source. Hydrogen and oxygen are in a liquefied state. Continuous stay under water is allowed for 21 days. Speed under water 20 knots.

The fuel cells installed on the Klass 212A submarines are capable of delivering up to 60-65% of the hydrogen energy.

"Platinum problem" is not limited to the price of metal for the catalyst. It also includes the problem of the frequency catalyst replacement due to its poisoning with impurities in fuel (energy carrier) and by-products of oxidation. The problem of poisoning by-products can be solved to some extent by selecting a reaction regime. The problem of poisoning with impurities in the initial fuel is solved either by deep purification of the fuel (which leads to a sharp rise in the cost of the process) or by switching to a process of producing fuel that does not create an admixture of catalytic poisons (which requires a change in the established technology - also with economic consequences).

Anyway, the development of serial fuel cells required the transition to more accessible raw materials for the production of catalysts. Relevant studies began actively in the 1990s, and led to practically meaningful results in the 2010s. These studies were logically aimed at finding alternatives among other metals of the VIII group of the periodic table.

Here it is appropriate to say a few words about the VIII group. Its main subgroup is inert gases, and the sub-group is metals, among which:

"Light" metals: iron, cobalt, and nickel.

"Heavy" (platinum) metals: ruthenium, rhodium palladium, osmium, iridium, platinum.

All of them have a certain generality of the quantum-electronic structure.

As a consequence of this generality, the "light" metals of Group VIII are similar to the "heavy" (platinum) metals of this group. This similarity determines the possibility of replacing catalysts based on platinum and palladium by nickel-based catalysts in the hydrogenation processes known through the work of Paul Sabatier in 1912. Paul Sabatier got Nobel Prize in Chemistry 1912 "for his method of hydrogenating organic compounds in the presence of finely disintegrated metals whereby the progress of organic chemistry has been greatly advanced in recent years." together with Victor Grignard [3].

Somewhat later, a number of catalytic activities of Group VIII metals were established in descending order for the reactions of hydrogenation of low molecular weight olefins: rhodium - iridium - ruthenium - platinum - palladium - nickel - iron - cobalt - osmium.

Modern investigations in the field of catalysis are focused, first of all, on organic complex compounds of "light" metals of group VIII. In particular, in a study of 2013 in the PNNL (Pacific Northwest National Laboratory), the goal was to reproduce the action of a natural catalyst - a hydrogenase enzyme that contains an iron atom, and activates hydrogen in living organisms. The result was a complex providing a voltage of 160 to 220 millivolts in a hydrogen cell: an indicator close to a cell on a platinum catalyst. Serial production of such catalysts based on iron instead of platinum are not yet reported, but the possibility of such production is beyond doubt.

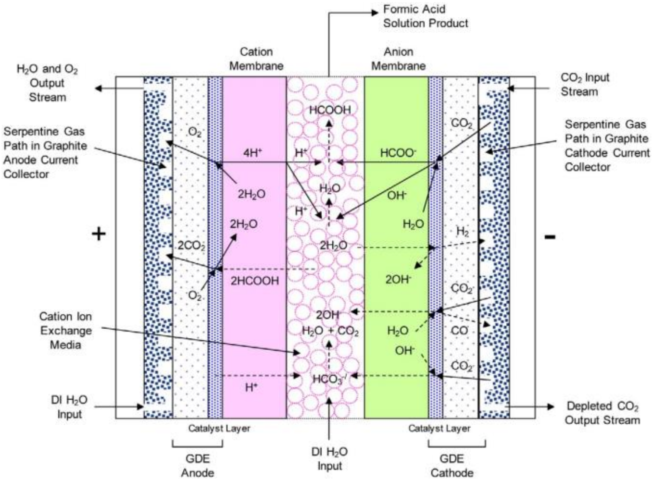

Fig. 1. Qualitative scheme of fuel cell produce formic acid [4].

The decomposition of formic acid with the evolution of hydrogen proceeds according to a similar scheme, so it was logical to assume the efficiency of the close-lying catalysts. This was confirmed in 2010. The group Matthias Beller from the University of Rostock published the result of work on the dehydrogenation of formic acid:

$$HCOOH \text{ -> } CO_2 + H_2$$

on a catalyst - a complex iron compound. This study is extremely important in the aspect of using formic acid as a hydrogen storage battery [5].

We will comment some results connected complex iron compound below. But here we will show quantitative efficiency of different Fuel Cell Technologies [6].

In general, all fuel cells have same basic configuration – an electrolyte and two electrodes. But there are different types of fuel cells, classified primarily by the kind of electrolyte used. The electrolyte determines the kind of chemical reactions that take place in the fuel cell, the temperature range of operation, and other factors that determine its most suitable applications

TABLE I: COMPARISON OF FUEL CELL TECHNOLOGIES

| Fuel Cell Type | Operating Temperature | System Output | efficiency | Applications |
|---|---|---|---|---|
| Alkaline (AFC) | 90-100˚C<br>194-212˚F | 10kW-100kW | 60-70% electric | Military<br>Space |
| Phosphoric Acid (PAFC) | 150-200˚C<br>302-392˚F | 50kW-1MW (250kW module typical) | 80-85% overall with combined heat and power (CHP) (36-42% electric) | Distributed generation |

| | | | | |
|---|---|---|---|---|
| Polymer Electrolyte Membrane or Poton Exchange Membrane (PEM)* | 50-100°C<br>122-212°F | <250kW | 50-60% electric | Back-up power<br>Portable power<br>Small distributed generation |
| Molten Carbonate (MCFC) | 600-700°C<br>1112-1292F | <1MW (250kW module typical) | 85% overall with CHP (60% electric) | Electric ability<br>Large distributed generation |
| Solid Oxide (SOFC) | 650-1000°C<br>1202-1832°F | 5kW-3MW | 85% overall with CHP (60% electric) | Auxiliary power<br>Electric ability<br>Large distributed generation |

## III. ELECTROCHEMICAL FORMATE SYNTHESIS: PROCESSES AND CRITERIA

Electrochemical preparation of formic acid by the method of $CO_2$ reduction at the cathode has been known for a long time. The first published study: Nikolay Beketov, "Formation of formic acid in the electrolysis of bicarbonate sodium," 1869. The process can be realized at different cathodes of different composition, and under different conditions.

The most interesting is optimization in three directions:

1) The maximum intensity of the process is the maximum yield of formic acid from the unit area of the cathode.
2) Minimum energy consumption - the process at a low temperature (below 100°C ).
3) Reversibility of the process - the possibility of creating a battery that in the charging mode restores $CO_2$ to formic acid, and in discharge mode - oxidizes formic acid to $CO_2$, using one catalytic electrode as the cathode in the charging phase, and the anode - in the discharge phase.

The multivariance of electrochemical synthesis is one of the reasons why formic acid looks like a promising renewable energy source and a promising agent for hydrogen storage.

Some experiments in [7] were performed with hydrogen generation from formic acid and direct application in a LT PEMFC including heat exchange. In Fig. 2 results of usage catalyst with iron show achieved periods of stable operating which is one of more important characteristics of the process.

The second major reason is the high current density in the fuel cells on formic acid. It is much higher than the current density for a similar oxidation of alcohols. Although it should be noted that the energy density for formic acid is several times lower (5.6 KJ / g vs. 22.7 KJ / g in methanol).

The third reason is the additional use of formic acid as a starting material for organic synthesis chains. For this reason, it makes sense to dwell in more detail, noting in advance that formic acid is both a source of hydrogen and source of pure carbon monoxide-II, CO, which is released by the reaction:

$$\mathrm{HCOOH} \rightarrow \mathrm{CO} + \mathrm{H2O}$$

in the presence of dehydrating agents.

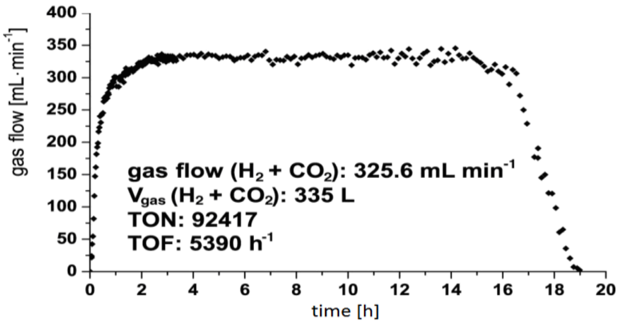

Fig. 2. Results of work [5] on time of stable catalytic process with iron usage.

## IV. FORMAT CYCLE IN OBTAINING CHEMICAL AND FUEL PRODUCTS

At present, several new processes already exist, analogous in a technological sense to photosynthesis realized by plants. The processes are realized in electrochemical cells, directly connected with solar panels, as sources of power supply. At a cell temperature of about 70°C due to a thin selection of catalyst, a specific yield of formic acid or products of its further reduction per unit of absorbed solar energy is achieved, exceeding the specific yield of organic in natural photosynthesis [7]. You can note the company «Liquid Light», founded in 2009 in New Jersey, and focusing exclusively on the development of such processes. A unit with similar themes was created in the corporation "Panasonic", and groups with similar themes are organized in several universities of America, Europe, and South Korea.

At the same time, relatively simple and economically successful chemical-technological methods are existing, known collectively as the "Fischer-Tropsch process" (FT process), have not been recalled. The FT-process was proposed in the 1920s in Germany [8].

At that time, synthesis gas - a mixture of CO and $H_2$, obtained by high-temperature coal treatment with coal - was already known. As shown by Fischer and Tropsch, the reactions of CO and $H_2$ on the catalyst of the "light" metals of Group VIII at a temperature of about 200° C and moderate pressure yield a mixture at the output that is close in physical and fuel properties to the gasoline fraction of the oil. This product FT-process was called "syntin" or "ersatz-gasoline". Relative simplicity and unpretentiousness of technology, which does not require any expensive and scarce materials, was in demand on the eve of the Second World War. In the 1940s, according to the FT-process, the production of synthine reached 6 million tons per year. The FT process showed significant variability when changing the conditions and the catalyst. This allowed to produce also more than 70 different products of organic chemistry. In the post-war era, the FT process was occasionally in demand in the oil crisis, and was modified in the part of product management. Today, clones of the FT process remained in Malaysia (the Shell plant) and South Africa (the PetroSA plant), but they are more focused on obtaining heavy paraffins from natural gas after pyrolysis.

Actually, loss of interest in the FT process since the 1990s has been associated with the expansion of the natural gas market as a convenient and widespread energy carrier. A number of initiatives in the field of man-made fuels have been discontinued for this reason, however, changes in the scope of control over the volume of combusted hydrocarbon fuel (including the Kyoto Protocol, $CO_2$ emission quotas, etc.) return the relevance to the FT process - in the case of its restructuring under the scheme with a "zero carbon balance". If the feedstock of the FT process (synthesis of gas: $CO + H_2$) is produced by the decomposition of formic acid, which is derived from atmospheric $CO_2$ by fixation using renewable non-carbon energy sources, the "zero carbon balance" is preserved.

In the context of this paper, the FT process is interesting in its original version: a simple technological production of ersatz gasoline on cheap catalysts. The facts are that, in spite of the extremely significant funds invested in the transition from cars with gasoline internal combustion engines to electric vehicles or other "green cars", this transition, even if ever, will certainly not in the coming decades. Thus, there is a clear economic prospect of using formic acid not only as an energy source for fuel cells, and not only as a hydrogen storage battery for "hydride" vehicles, but also as a combined $H_2$ and CO accumulator for obtaining traditional fuel for cars which does not entail a violation of $CO_2$ emission quotas.

Next, we will pay attention to two questions related to the content of the main part of the article. One of them is the possible expansion of the use of formic acid. The second question is related to the concept of global warming, which greatly affects the content of work on energy development.

## V. USE OF THE PROPOSED DIRECTION FOR THE BINDING OF $CO_2$ IN CONJUNCTION WITH THE DISPOSAL OF CERTAIN TYPES OF NON DISPOSABLE INDUSTRIAL WASTE

Formic acid synthesized by electrochemical method from atmospheric $CO_2$ in aqueous solution can be used as an energy carrier for a fuel cell, not in the form of HCOOH itself, but in the form of a salt (format) of a metal.

In particular, iron or other metal formats obtained by washing secondary metal (large-capacity scrap) from corrosion layers (oxide or hydroxide of metal) can be used. When oxidizing formate in the fuel cell, in this case the reaction occurs:

$$HCOO(-) - 2e + H2O = CO_3(2-) + 3H(+)$$

Formats of transition and alkaline earth metals are soluble in water. The carbonate-ion formed by the oxidation of formate carbonate $CO_3$ (2-) forms carbonate-insoluble carbonates with the same metals. Those. after the fuel (formic acid, formate) is processed in the fuel cell, the oxidation product, i.e. $CO_2$, does not return to the atmosphere, but settles in the reaction vessel and can be separated as a solid phase.

Carbonates of iron and other metals are practically non-toxic, fairly stable, and can be added to any non-acidic building mixes (concrete, sintering solution for ceramics, etc.) - in this case, $CO_2$ is fixed for an indefinite time in the building material. In addition, it is possible to use the property of transition metal carbonates to decompose with the release of $CO_2$ at a temperature of about 500 ℃. Those. It is possible to use carbonates as foaming agents in the production of foam ceramics. In this case, the gaseous $CO_2$ remains fixed in the foam (deaf pores) of the foam ceramic product. Thus, it is possible to use formic acid in two ways depending on the state of energy supply problems and the reduction of $CO_2$ emissions, if the urgency of this task is maintained.

## VI. SOME CONSIDERATION REGARDING THE DEGREE OF PERSUASIVENESS OF THE GLOBAL WARMING DOCTRINE FROM THE IMPACT OF TECHNOGENIC GREENHOUSE GASES

The processes of climate change are extremely complex, not yet fully investigated, depend on a number of factors, and have a complex periodicity with several amplitude-frequency characteristics. On intervals of the order of centuries these processes are subject to significant fluctuations. Climate monitoring is conducted during a historically short period, in which it is statistically difficult to separate systemic and random changes. The last remark is due to the fact that experts in climatology and earth physics are reasonably saying that the warming period will soon end. Below we give two quotes of famous experts in climatology.

"In fact, we now live at the peak of a relatively small local warming, after which a new phase of cooling will begin (if it has not already started). The main regulator of the temperature of the earth's atmosphere is the radiant energy of the Sun, connected with the distance between the Earth and the Sun. The distance varies periodically with time. A few years ago it was minimal, which led to a warming, and now it starts to increase. In 10-15 years, not global warming awaits us, but on the contrary - global cooling ". Gorodnitsky [9].

"And every time it was warming that preceded the increase in the concentration of $CO_2$ in the air. The theory of the greenhouse effect assumes the reverse sequence. Cosmic factors lead to serious climate changes. The temperature changes the fluctuations in solar activity, as well as changes in the slope of the earth's axis, the period of revolution of our planet. Such fluctuations in the past, as we know, led to the onset of glacial periods" A. Kapitsa 2001 [10].

The history of the planet includes periods of hundreds of millions of years, during which the concentration of carbon dioxide was an order of magnitude higher than now. So, in the Cambrian period, it was about 20 times higher than today. Then, during the Paleozoic era, it was roughly equal to modern or even lower. After that, in the Mesozoic era, it was about 10 times higher than the modern one.

In this case, neither in the Cambrian period. Neither in the Mesozoic period, there was an irreversible greenhouse $CO_2$ effect with the heating of the planet by analogy with the greenhouse processes on the planet Venus (closer to the Sun). The reasons for the lack of such warming up for the Earth has a well-known calculation-physical explanation:

"A direct calculation shows that the greenhouse effect caused by carbon dioxide plays a very limited role in the heating of the earth's surface. The amount of carbon dioxide in the earth's atmosphere is 500 times less than water vapor, and the windows of non-transmission of these gases in the infrared part of the radiation overlap. In addition, the greenhouse effect of carbon dioxide is masked by the screening effect of cloudiness in both directions of radiation propagation" [11].

The doctrine of global warming under the influence of technogenic greenhouse gases does not contain clear answers to these comments - at least for now. Given this situation, work that is inextricably linked to especial on this doctrine should perhaps be treated with reasonable caution.

One of well-known specialist on struggle with $CO_2$ emission is R. Allam [12].

## VII. SOME FINAL GENERAL CONSIDERATIONS

In conclusion, it is appropriate to cite some of the theses related to "green" energy as applied to the formate cycle considered in this paper.

To date, the "green" energy industry is developing in two ways.

1) Small "green" energy - providing energy from renewable sources (sunlight, wind energy, and tidal waves) of small objects for which the power consumption is less than 2 MW (apartment building or small settlement).
2) Large "green" energy - direct replacement (or, more accurately, an attempt to directly replace) large power plants of the same power "green" stations - wind and solar farms.

In the first way, significant progress has been made, which is natural, since at this level of power consumption, "green" energy sources are easily integrated into the structure of an apartment building - condominium, or a small village. Based on the typical specific production of "green" solar or wind energy: 20 MW / km$^2$, the production of 2 MW requires 10 hectares of area - the residential complex usually has such an area, taking into account the possibility of placing "green" units on the roofs, technological household elevations. In addition, there is no problem of long-distance transmission of generated electricity - it is consumed locally. Here the problem is only in the accumulation of electricity for food during the peak consumption period or during the period of low power removal of the "green" source. This is a problem that can be solved with the use of formic acid as a storage energy source.

## VIII. CONCLUSION

Formation technologies (based on a renewable resource - atmospheric $CO_2$) and positively affecting the fight against the so-called. "Greenhouse gases" and "global warming" can partially replace the oil and gas technologies that dominate modern industrial organic chemistry.

The article also gives some results of the study of the use of different versions of catalysts for increasing the efficiency of hydrogenation and dehydrogenation processes in fuel cells.

When implementing synthesis processes, the production of formic acid from atmospheric carbon dioxide (or from the carbon dioxide emissions of industrial or energy enterprises) has prospects as a separate ecological branch of industrial organic chemistry - the so-called. «Green technologies».

The most important part of the concept is the prospect of widespread use of the well-known Fischer-Tropsch process, which has lost relevance due to the huge growth in oil and natural gas production.

Here it is also necessary to note the following. The scale of the practical use of the ideas presented is closely related to the dynamics of the transition to new energy generation schemes. In particular, this applies to mobile power plants (mobile electric generators on fuel cells). Energy consumption in the schemes in question occurs without increasing the amount of $CO_2$ in the atmosphere, and the combined waste management scheme provides additional absorption of $CO_2$.

## NOMENCLATURE

| | | |
|---|---|---|
| FA | Formic Acid | |
| FT | Fischer-Tropsch process | |
| EPFL | Ecole Politechnique Federale de Lausanne | |
| TON | Turnover Number | «-» |
| TOF | Frequency of Turnover | 1/h |

## CONFLICT OF INTEREST

The authors declare no conflict of interest.

## AUTHORS CONTRIBUTIONS

M. Gotovsky has written most part of the paper and supervised the research conduction. A. Gotovsky was the author of main ideas of the chemical part of the paper. V. Mikhailov and Yu Sukhorukov analysed the method of data obtain and supervised energetic part of the paper. V. Lychakov and E. Sukhorukova conducted necessary studies.

All authors had approved the final version.

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