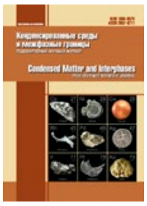

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Prospects of membrane catalysis in hydrogen energetics. Mini review

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Abstract

Hydrogen energetics is undoubtedly highly relevant today as it not only allows solving the issue of energy production from a renewable water source but can also prevent the formation of greenhouse gases. They say that any new idea is a well forgotten old one. The paper is dedicated to an excellent but still unimplemented work of Sainte-Claire Deville who managed to obtain hydrogen from water vapor using membrane technology. He used a clay pipe as a membrane which selectively permeated hydrogen. This process occurred with heating up to 950 °C. Sainte-Claire Deville managed to obtain only a mixture of hydrogen and oxygen in a ratio of 4:1 and then to clean the product from oxygen using chemical reactions.

Modern membrane catalysts based on palladium or its alloys are selectively permeable only for hydrogen. This means that the membrane catalysis method with palladium membranes could allow to realize of thermal water disassociation more effectively and solve the issues of hydrogen energetics using only renewable raw materials.

The history of hydrogen discovery and methods of its production was also studied in this review. Different methods of energy production were analyzed, including mineral resources, wind turbines, solar panels, hydroenergetics, electrolysis, and nuclear power, and a forecast was presented based on them. The review should be considered as an invitation to further discussions regarding this highly relevant and important topic.

Keywords: Energy, Hydrogen, Ecology, Economy, Sustainable development, Membranes, Palladium, Catalysis, Water vapor

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1. Introduction

1.1. Early history of hydrogen energetics

There is no future yet. The present is the future that continually drifts into the past. Therefore, only the past is real, and prediction is also an action related to the past.

The discovery of hydrogen by Henry Cavendish in 1766 [1] was the preface to the history of hydrogen energetics, although Cavendish himself believed that he had discovered phlogiston, a combustible gas that his contemporaries had been looking for. To obtain hydrogen, Cavendish used the reaction of sulfuric or hydrochloric acid with zinc, iron, or tin. He discovered that hydrogen burns in the air. This method is now used in schools and for first-year chemistry students but it is not used for industrial production due to the high cost of reagents. The history of hydrogen energetics began in 1880. When Anthony Carlisle, professor of medicine from London, learned about the structure of the voltaic cell, he cancelled his lectures, postponed the surgeries, and produced the element using 17 silver half crowns and 17 zinc plates. Then he put the wires soldered to the outermost plates into water and observed the outgassing. William Nicholson helped him establish that one electrode released hydrogen and the other one released oxygen. The volume of the released hydrogen was twice more than the volume of the released oxygen. Having satisfied his curiosity, Carlisle returned to his previous work while Nicholson and Cruikshank continued the study of the electrolysis of solutions of salts and published their results [2].

Fig. 1. Sainte-Claire Deville (1818-1881)

Henri Sainte-Claire Deville suggested a new approach to the issue of hydrogen energetics (Fig. 1) [3–6]. Deville's array is presented in Fig. 2. Its main component is a pipe made from annealed non-glazed kaolin. Kaolin is an aluminosilicate $Al_2O_3 \cdot 2SiO_2 \cdot 2H_2O$ with a porous structure and it has the properties of a weak cation exchanger. A kaolin pipe was placed in the middle of the furnace, and water vapor was supplied to the pipe through valve *∂* from a flask or a retort. The kaolin pipe was inserted into a wider glazed porcelain pipe that was gas-impermeable. The edges of the pipe were sealed with clay paste. The pipes were placed in the furnace that was stoked with small pieces of coke. It was established that a

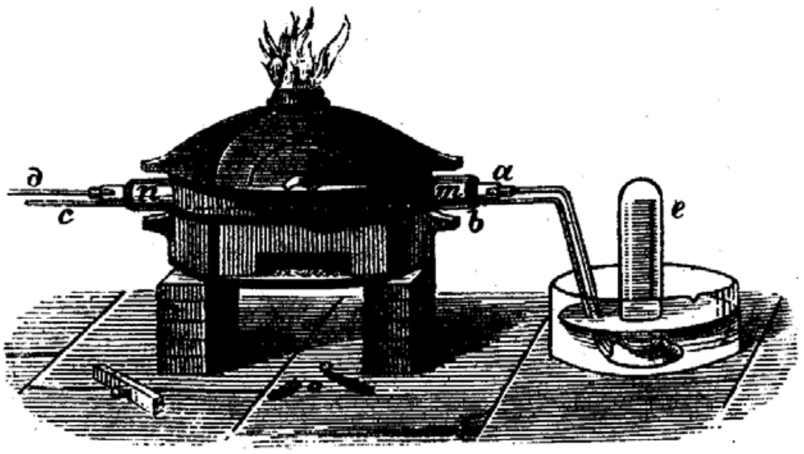

Fig. 2. Sainte-Claire Deville's array for the production of hydrogen

temperature of 950 °C is enough for the thermal disassociation of water molecules.

Sainte-Claire Deville was familiar with the works of Thomas Graham who studied membrane processes and founded colloid chemistry [7]. Graham showed that the flows of hydrogen through glass membranes or animal membranes are 4 times more than the flows of oxygen. The result obtained by Deville upon the diffusion through a kaolin membrane was similar to the result obtained by Graham upon the diffusion of hydrogen and oxygen through other kinds of membranes. Thus the space between the kaolin and porcelain pipes was enriched with hydrogen but still contained oxygen. To prevent the mixture of hydrogen and oxygen from burning, Sainte-Claire Deville passed carbon dioxide through pipe *c* between the pipes through the ring-like space. The rest of the oxygen reacted with hydrogen with the formation of water. Carbon dioxide that was released from pipe *b* was neutralized by alkali. As a result, at the outlet of the space between the pipes, Deville obtained pure hydrogen. A clay membrane is not highly selective in relation to hydrogen. Today there are membranes that permeate hydrogen selectively, which can significantly improve Sainte-Claire Deville's method. It should be noted that if there is no selective membrane upon thermal disassociation, even at the pressure of 0.1 at. 40 % the disassociation can be achieved only at 3000 °C.

1.2. Analysis of modern methods of hydrogen production

In past centuries, technologies were evaluated only from an economical perspective. Due to the challenge thrown down to us by nature, it is now necessary to assess technologies not only from an economical, but also from an environmental perspective. Above all, we are talking about the deviation of the changes in the Earth's surface temperature from a cyclic dependence [8] towards an exponential increase over time [9]. It is considered to be global warming. It is caused by the increased concentration of greenhouse gases. Up until a certain concentration, greenhouse gases are necessary to keep warmth in the atmosphere but their excess results in global warming, the signs of which are already apparent. Up until a certain concentration, greenhouse gases are necessary to keep warmth in the atmosphere but their excess results in global warming, the signs of which are already apparent. As a result, the generation of greenhouse gases and primarily carbon dioxide should be taken into account when evaluating new and existing technologies.

The words economics and ecology have the same root. It is a Greek word οίκος which means home. In this case, it is the Earth, our home. However, despite the tendency towards globalization, economics is mostly of local nature, as countries are protected from one another by their borders, while the environmental situation is fundamentally global, as convection tends to equalize the gradients of temperatures and concentrations. Therefore, environmental issues are worldwide and cannot be nation-specific.

Let us discuss the issues of hydrogen energetics from this point of view. Industrial progress begins with science. Scientific ideas are implemented when a fertile field is provided for them. Some issues relating to implementation require additional research. This is how a close connection between science and production is formed. An example of this is Justus von Liebig's discovery of the irreversible extraction of minerals from soil for human nutrition [10]. He suggested using mineral fertilizers containing nitrogen, potassium, and phosphorus to preserve soil fertility. Potassium and phosphorus can be introduced into the soil as natural compounds while hydrogen is required in order to produce ammonia for nitrogen-containing fertilizers. BASF was the first to solve the issue of the production of ammonia for fertilizers. The researchers who worked on this project were awarded with a Nobel Prize (F. Haber and C. Bosch) [11]. A. Mittasch discovered an effective and cheap catalyst to synthesize ammonia from nitrogen and hydrogen. Large-tonnage production of ammonia was first organized in 1920 at a Leuna enterprise, not far from Merseburg. Nitrogen and hydrogen are required for the synthesis of ammonia. And while nitrogen is the main component of atmospheric air, hydrogen can be obtained through chemical technology methods. Thus the issue of large-scale production of hydrogen was encountered.

Today, hydrogen for the synthesis of ammonia is obtained through the reaction of methane with water vapor [13]. The reactor's tube is a membrane catalyst. Pd, Pt, Ru, Rh, and Ir [14] are the most efficient materials for the tube, although an Ni catalyst, which is significantly cheaper, is more often used in practice. Fe and Co, which are more reactive, are not used due to their oxidation during the reaction. A typical catalyst consists of relatively large particles of nickel deposited on Al_2O_3. The process is conducted at high temperatures (up to 1000 °C), but when using membranes made of palladium and silver alloys, temperature must be reduced to 500 °C. The reaction results in a mixture of H_2, CO, and CO_2. The reaction proceeds as follows:

$$\begin{aligned} &CH_4 + H_2O = CO + 3H_2 \quad \Delta H^o_{298} = +206\ \text{kJ/mol}; \\ &CO + H_2O = CO_2 + H_2 \quad \Delta H^o_{298} = -41\ \text{kJ/mol}. \end{aligned} \tag{1}$$

The method for the production of hydrogen for the synthesis of ammonia through the reaction of methane and water vapor is different from Sainte-Claire Deville's method that used only water vapor as a reagent. The use of the mixture of methane and water vapor is economically efficient. The profitability of methane for the production of hydrogen led to the idea of excluding water vapor from the reagents, but this method requires a non-renewable energy source that will become exhausted over the course of this century. Apart from that, an increased content of methane leads to the deposits in the form of curved graphite layers on the surface of the carbon catalyst [14]. The formation of threads from them is similar to the synthesis of carbon nanotubes. As a result, the reactor becomes blocked and the catalyst is destroyed. The introduction of water vapor into a reaction phase prevents the deposit of carbon on the surface of the catalyst. As a result of the experiments with various ratios of water vapor and methane, it was established that an increase in the proportion of water vapor leads to an increase in the proportion of methane that is converted over the course of the reaction [13]. Sulphur that is present in natural gas not only poisons the catalyst, but also hinders the use of hydrogen in the fuel cell [14]. Reactions (1) show that apart from hydrogen, carbon dioxide, a greenhouse gas, is also produced, while carbon oxide, a poisonous gas, is an intermediate product. As methane, the main component of natural gas, is a non-renewable energy source, the improvement of methods and technologies of hydrogen production becomes a relevant task.

The method for the production of hydrogen by electrolysis from water or water solutions of salts has been known for a long time [2] and is still used today [15–20]. Electricity is required for electrolysis. Today, 37 % of electricity is generated by coal burning. The production of electricity by heat power plants seems to be environmentally friendly, but it gives the wrong impression, if we look at it from a consumer's perspective. To produce 1 kW of one hour of electricity by coal burning, 0.5 mol-Eq of sulfuric and nitric acids are released in the atmosphere, which leads to acid rain that destroys forests, damages architectural monuments, and increases metal corrosion. Apart from that, 1 kg of carbon dioxide (greenhouse gas), 14 g of slag, as well as radioactive isotopes are also released in the atmosphere [17–19]. The transition to the production of electricity from natural gas has considerably improved the environmental situation. Of all power plants, 24 % work on natural gas, although we should not forget that natural gas is a non-renewable energy source, and its exhaust product is carbon dioxide, a greenhouse gas [20].

The use of energy from the flow of rivers has been used for a long time, and now its proportion in the total production of electricity is 16 %. Although water-power is a renewable source of energy, there are a number of obstacles that prevent this method from being utilized more. Dams must be built in order to construct water power plants, which leads to changes in natural conditions. Water above the dams floods land that can be used for agriculture. And it also floods picturesque natural and historical places. Silt accumulates above the dams, while below the dams the concentration of nutritional substances in water decreases. Dams stand in the way of migration and the spawning of fish. Due to this, the USA is not planning to build new dams. Moreover, one dam in Maine has already been demolished, and they are going to demolish two more in Columbia [8]. Because of this, the future of hydroenergetics does not look optimistic.

Today, nuclear power stations produce 10 % of electricity. In some countries (USA, Russia, France, Japan, and others) nuclear power stations are considered to be the most promising source of energy. Nuclear power stations produce electricity constantly, 24 hours a day, and since many enterprises do not operate at night, during this time nuclear power stations can be used for the production of hydrogen by electrolysis. Modern thermal-neutron and slow-neutron reactors operate on enriched uranium-235, the proportion of which is only 0.7% natural uranium. Fast-neutron reactors can operate on uranium-238 the content of which is 99.3 %, which allows using stocks as reactor fuel. In Russia, an energy unit with a fast-neutron reactor has been operating at the Beloyarsk Nuclear Power Station for 30 years. Although this technology cannot be considered a completely renewable source of energy, still it can be said that it considerably expands the possibilities of obtaining a long-term energy source as compared to natural gas and oil. However, a number of countries (Germany, Italy, Spain, Belgium, Switzerland, and Austria) have decided against using nuclear power stations. It was due to the disquiet caused by the Chernobyl and Fukushima accidents. It is also possible that the leaders of these countries consider them to be dangerous in case of wars when nuclear power stations can easily cause a disaster. Today, the future of nuclear power stations cannot be definitely evaluated.

Wind power is a free, environmentally friendly, and renewable source of energy. Sailing boats and mills have been using wind power for a long time. At present, worldwide wind energetics is limited to 5 % of all power production, but European researchers and engineers are improving wind engines and are planning to have this figure increase up to 20 % by 2030. However, there are some issues related to the use of wind power. First of all, among them are the intermittent nature of the process and the transfer of energy over long distances. There are also some difficulties associated with the installation of wind generators, and wind turbines may freeze in winter. These issues do not prevent wind energetics from developing, but it is obvious that it will not be able to become the dominant source of energy.

Another relevant issue is how to increase the efficiency of solar energy. Currently, it claims only a tiny portion of the worldwide production of electricity (3 %). There have been both successes and failures in this area. J. Carter, a former president of the USA, was keen on the idea of solar energy and had a solar heat collector installed on the roof of the White House, but there was a leak in the collector and it was then placed in the Jimmy Carter Library and Museum. Not all regions are effective for the use of solar energy, so, as part of a new project, photocells were placed in the Sahara Desert and the energy was later transferred to Europe.

As a result of the analysis, we can conclude that the production of hydrogen through the membrane catalysis of water vapor and the use of nuclear power stations for the electrolysis of water or water solutions of salts with the further use of fuel cells to create power stations are promising for energetics [20].

2. Membrane catalysis for hydrogen production

The term sustainable development was suggested in 1983 when UN assembled the World Commission on Environment and Development. The concept of sustainable development has become a paradigm and it should be understood as development that meets today's requirements without affecting the ability of future generations to satisfy their own needs. From this point of view, the present intensive use of hydrocarbon fuel and coal deprives future generations of energy sources. Thus, the method of membrane catalysis that uses water vapor as a reagent seems to be the most promising as it involves a renewable energy source and it is already free from harmful emissions into the soil and atmosphere.

The implementation of the method of membrane catalysis does not require any fundamentally new devices. The units that are used for hydrogen production from ammonia synthesis can be used for the industrial implementation of this method. To start using the new technology, one just has to stop using methane in the reforming processes and use materials with nanodispersed palladium on the surface as membranes. Palladium will act as a catalyst in this process.

The electronic structure of palladium in increasing energy is $1s^2\ 2s^2\ 2p^6\ 3s^2\ 3p^6\ 4s^2\ 3d^{10}\ 4p^6\ 4d^{10}$. As a result, the palladium membrane selectively permeates only hydrogen, which can

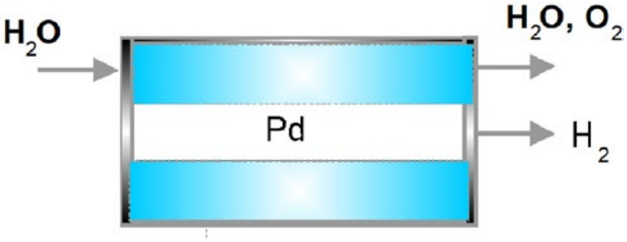

Fig. 3. Reactor for obtaining hydrogen from water vapor

be used as a source of energy or be converted into electrical energy using a fuel cell.

The exceptional selectivity of the palladium membrane can be explained from the perspective of the electrostatic interaction of its surface with water molecules. Fig. 4 shows an interaction model based on the example of a palladium atom and a water molecule. On the left is the initial state in which the water molecule is on the surface of the palladium. The water molecule has a constant electric dipole moment $\mu = 1.84\ (\pm\ 0.02)\cdot 10^{-18}$ el. un., see [19]. When determining the value of the dipole moment, the direction from the negative pole of the dipole to the positive one is conditionally introduced. The values of quadrupole and octupole moments of water also provide useful information on the redistribution of charge in a molecule. Each atom is also a dipole with a positively charged core and negatively charged mobile electron shell. Cores and electrons oscillate, and as a result, potential energy is conditioned by the electrostatic interaction between dipoles. It should be noted that the surface of any atom, and especially palladium due to its specific electron structure, is negatively charged, which is highly important for understanding the mechanism of any catalytic process. As the water molecule is strongly polar, its hydrogen atoms are oriented towards the negatively charged surface of palladium. The conducted quantum chemical calculations show the result of the interaction [21] where the water molecule was broken by Coulomb forces and the hydrogen atoms were inside the palladium membrane, while the oxygen atom remained on the membrane's surface. The hydrogen atoms that passed through the palladium membrane due to the chemical potential are recombined into molecules on the opposite side of the membrane. Membrane catalysis is different from surface catalysis because reagents are adsorbed on the surface and after the reaction its products are detached from it. For example, a strong triple bond in the nitrogen molecule breaks on the catalyst surface during of the synthesis of ammonia, and then the nitrogen atoms react with hydrogen atoms and detach from the surface when ammonia molecules are obtained. The adsorption of reagent molecules on the catalyst surface is also a part of the membrane catalysis, but after that, one of the products diffuses through the membrane. This contributes to a more complete separation of products and reagents. This is the advantage of membrane catalysis.

The presented option for hydrogen production by membrane catalysis for its further use as a fuel or a component of a fuel cell is just one example of the global processes occurring in the universe. First of all, we use the energy of the sun directly or indirectly. At high temperatures, hydrogen atoms are ionized and turned into cores (protons) on the sun and other stars. The source of energy of stars is a chain of reactions, the main of which is the

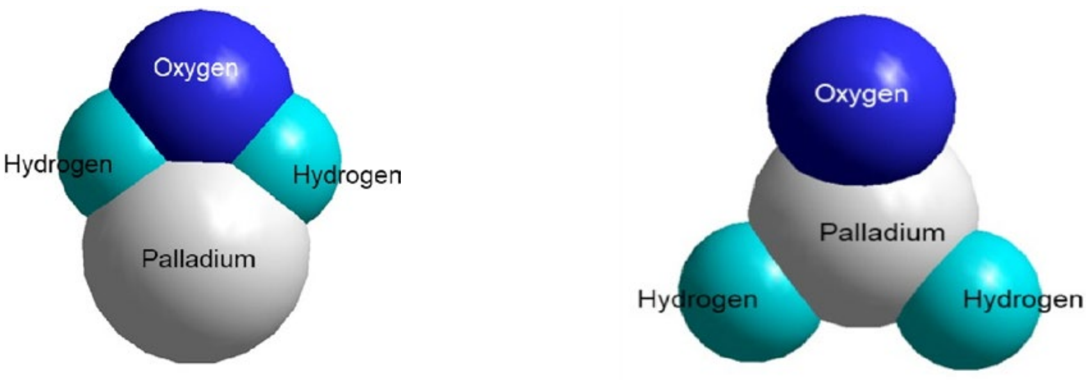

Fig. 4. Interaction model of water molecules and palladium atom. On the left: before the beginning of the catalytic reaction, on the right: during the reaction

reaction between the hydrogen cores the result of which is helium:

$$4\,{}^{1}_{1}H^{+} \rightarrow {}^{4}_{2}He^{2+} + 2e^{-} + 26.7\ \text{MeV}. \qquad (2)$$

The sun is comprised of 75 % hydrogen and 24% helium. The interstellar medium is comprised of 90 % hydrogen and 10 % helium, which corresponds to the elemental composition of young stars. The lifetime of our planet is approximately 5 billion years and we are now in the middle of its life.

On average, only about 0.3% of all solar energy that reaches the surface of the Earth is captured in the course of photosynthesis. Chemists have set themselves the goal of synthesizing supermolecules that would allow the absorption of solar energy to be considerably increased, but no prominent results have been achieved so far [22]. According to A. Szent-Gyorgyi, a Nobel Prize laureate, the process of photosynthesis is primarily a reaction that uses solar energy for the disassociation of water:

$$nH_2O + nCO_2 \xrightarrow{h\nu} C_nH_{2n}O_n + nO_2, \qquad (3)$$

while the obtained hydrocarbons, in his opinion, are similar to a hydrogen supply, which is mainly used to generate energy through its burning [23]:

$$\begin{aligned} &C_nH_{2n}O_n + nH_2O = 4nH + nCO_2; \\ &4nH + nO_2 = 2nH_2O + \text{Energy}. \end{aligned} \qquad (4)$$

He believed that living organisms contain only one type of fuel, hydrogen.

Photosynthesis produces a great amount of biomass that can be used as a renewable source of energy. In Brazil, ethanol produced from biomass amounts to 30-60 % of car fuel. Up to now, people have been extracting hydrocarbons that were obtained from the products of photosynthesis in the depths of the Earth millions of years ago. However, it is possible to transform the products of photosynthesis into a source of energy, right here and right now. Bioenergetics also has some issues. Today, ethanol is more expensive than gasoline, but it is not the only thing that prevents us from increasing the use of biomass. The main problem is that more space is needed for the production of biomass, and this comes into conflict with the need for more space for food crops due to the exponential growth of the population. The use of algae seems to be promising for this purpose as they can provide three times more biomass per unit of space than palm plantations and six times more than corn plantations.

3. Conclusions

The analysis of the methods of energy production allowed determining the most promising methods for future hydrogen energetics. The first of them is energy from nuclear power plants that can be used for the production of electrolytic hydrogen. The second one is the use of ethanol obtained from biomass by photosynthesis. The third and the most promising one is the production of hydrogen by membrane catalysis through thermal disassociation of water vapor. A metal with palladium deposited on its surface that selectively permeates hydrogen can be used as a membrane. Hydrogen can be used as a fuel without any harmful emissions. However, hydrogen can also be used in a fuel cell in order to produce electricity. This is how we see the future of power plants. In some cases, the nuclear power plant energy can be used for the heating of a membrane reactor.

Conflict of interests

The author declares that they have no known competing financial interests or personal relationships that could have influenced the work reported in this paper.

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