Promising methods for hydrogen producing

G.E. Ergazieva[1,3], M.M. Telbayeva[1,2], K. Dossumov[1], A.I. Niyazbayeva[3]

[1]The Institute of Combustion Problems, Bogenbaybatyr str.172, 050012, Almaty, Kazakhstan
[2]Kazakh state women's teacher training university, Aiteke bi str. 99, 050000, Almaty, Kazakhstan
[3]al-Farabi Kazakh National University, al-Farabi av. 71, 050040, Almaty, Kazakhstan

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ABSTRACT

Hydrogen production is one of the most promising ways to develop the energy sector of the future. Hydrogen does not exist in nature in its elemental form and therefore, it must be obtained from hydrocarbon, water or any other hydrogen-containing compounds. The variety of potential sources of raw materials for hydrogen production is one of the important reasons, in which hydrogen is such a promising energy carrier. The article describes methods for preparing the basic energy carrier, hydrogen from natural gas, ethanol etc. Among various types of raw materials, bioethanol is very attractive because of its relatively high hydrogen content, availability, as well as safety during storage and handling.

Keywords: hydrogen, methane, bioethanol, catalyst, conversion, raw materials, natural gas, green technology.

Introduction

In connection with the transition to «green» technology worldwide, intensive work is under way to search for alternative sources of energy and energy carriers [1]. One of the promising, modern energy carriers is hydrogen, which is determined by ecological purity, universality and high efficiency of energy conversion processes with its participation. Hydrogen is the cleanest of all existing fuels. When burning in pure oxygen, the only products are high-temperature heat and water. The first place among the energy carriers in terms of heat of combustion is hydrogen. Hydrogen also use as a raw material in organic chemistry, petrochemistry, oil and gas processing. In the chemical industry, hydrogen serves as the main intermediate in the production of ammonia, methanol, synthetic fuels, with deep processing of oil and the production of high-octane motor fuel. Hydrogen use in low-tonnage, science-intensive industries: electronic, pharmaceutical, food, metallurgy, the synthesis of chemically highly active substances and other industries. Consumption of hydrogen all over the world is steadily increasing.

To date, it is relevant to use hydrogen as an energy carrier. Special attention was paid to the development of hydrogen energy in the mid-20th century at the height of the first wave of the energy crisis. It was based on the idea of hydrogen as an alternative to clean fuel, whose share in the fuel and energy complex was supposed to be commensurate with the specific gravity of organic fuel. In the last century, at the world level, many scientific events devoted to hydrogen were organized and held, where the participants expressed the opinion that the widespread use of hydrogen in power «provides humanity a unique chance to survive in a world free from environmental and social disasters» [2]. The first International Conference on Hydrogen Energy was held back in 1976 in the United States, which called for the use of hydrogen in power engineering and accelerated scientific research and development to produce hydrogen on an industrial scale. In 2008, in the United States, the majority of vehicles (about 90%) moved on compressed hydrogen gas, the remaining 10% – on liquid hydrogen. In addition, the predominant part of the cars (about 90%) was equipped with electric motors, the electric energy for which was produced by hydrogen fuel cells. At the end of 2008, there were 61 hydrogen filling stations in the USA [3]. In 2013 Toyota (Japan) released a new car Toyota Mirai, working on hydrogen. A special fuel cell that produces a chemical reaction due to hydrogen and oxygen in the car, converts electricity, which is the driving force of the machine. The only by-product of this process is water [4].

Hydrogen in the Free State on Earth is absent. The traditional method of obtaining hydrogen is the electrolysis of water, but one of the obstacles to large-scale use of this method to this day is the large consumption of electric current. To obtain hydrogen, available chemical raw materials are needed.

**Ответственный автор*
E-mail: telbaeva.moldir@yandex.kz (M.M. Telbayeva).

2 The methods for obtaining of hydrogen

Many of the substances occurring in nature contain hydrogen. One of them, water, which naturally occurs in the form of sea water, rivers, rain or well water, is the most widespread. Hydrogen can also be extracted from fossil hydrocarbons, biomass, hydrogen sulfide or some other substances. When hydrogen is extracted from fossil hydrocarbon, all carbon dioxide must be recycled (separated, trapped, etc.). In this way, in order to were not released greenhouse gases or other pollutants into the atmosphere, and the extraction process of hydrogen can be called «green». A variety of methods for producing hydrogen is one of the main advantages of hydrogen energy, as much us increases energy security and reduce reliance on individual types of raw materials. These include natural gas, water electrolysis, gasification, steam methane conversion, photocatalysis, bioethanol conversion.

2.1 Electrolysis of water

Probably, in the near future, methods for producing hydrogen using carbon raw materials would be the primary. However, the raw materials and environmental restrictions of some hydrogen production processes stimulate the development of hydrogen production processes from water. Among the methods for producing hydrogen from water, electrolysis, thermochemical and thermoelectrochemical cycles are the most interesting in the context of atomic-hydrogen energy.

Electrolysis – the decomposition of water using electric shock:

$$2H_2O + 4\,e = 2H_2 + O_2 \quad (1)$$

Electrolytic hydrogen is the most affordable, but an expensive product. In the world produced of electrolyticallyonly 4% of hydrogen. For the decomposition of pure water at ambient conditions required voltage of 1.24 volts. The voltage depends on the temperature and pressure and the properties of the electrolyte and other elements of cell.

The most attractive features of electrolysis technology are environmental (of course, on condition that the production of primary energy is not associated with environmental pollution), the possibility of establishing plants with widely varying performance (from a few litres to hundreds m^3 of hydrogen per hour), simplicity of operation and convenience in work, the high purity of the hydrogen produced and the presence of a valuable by-product - gaseous oxygen.

Basically, an aqueous electrolyzer contains two half-reactions: hydrogen providing reaction and oxygen providing reaction [5]. For these processes, electrocatalysts based on noble metals were widely used, including Pt, Ir, RuO_2, etc. Accordingly, practical application was seriously made difficult by their deficit and high cost. As a rule, the oxygen providing reaction include a slow 4e transfer process, the associated reaction mechanism and the paths are more complicated than hydrogen providing [6].

Therefore, despite the environmental effectiveness this method of producing hydrogen, the process remains expensive in the application noble metals and high energy consumption.

2.2 Direct thermolysis of water

One-stage thermal dissociation of water is known as thermolysis of water and can be represented as:

$$H_2O + 0.5O_2 \quad (2)$$

When heated above 2500 °C water is decomposed into hydrogen and oxygen (direct thermolysis). Such high temperature can be obtained, for example, using solar energy concentrates. One of the disadvantages of this process is the need for an effective method for the separation of H_2 and O_2, in order to avoid the ingress of an explosive mixture. Semi-permeable membranes based on ZrO_2 and other high-temperature materials can be used for this purpose in temperatures up to 2500 K (2226, 85 °C). Separation can also be achieved after the product gas mixture is cooled to a lower temperature. Baykara et al. [7] experimentally investigated the thermolysis of water using solar radiation. The authors concluded that the recombination of hydrogen and oxygen can be avoided by drastically lowering the temperature by 1500-2000 K (1226,85-1726,85 °C) for several milliseconds. But, for more efficient separation of hydrogen, further processes with the using palladium membranes will be required. Therefore, the process remains economically inefficient.

2.3 Gasification

Gasification: decomposition of hard hydrocarbons and biomass to hydrogen and gases for further reforming. Gasification - conversion of the organic part of solid or liquid fuel into the combustible gases at high temperature (1000-2000 °C) heating by co-oxidant (oxygen, air, water vapor, CO_2 [8], or, more likely, their mixture).

Disadvantages of the seprocesses that they take place at high temperatures.

Currently, most of the hydrogen produced in the industrial scale in the world is obtained by the steam reforming of methane (SRM) in catalytic processes. Kazakhstan has large reserves of energy resources, the country has large reserves of concentrated methane at Kumkol, Karachaganak gas fields, in the composed of natural and associated petroleum gas, as well as in coal seams of the Karaganda basin. However, currently the methane is used mainly as a fuel (home and car) or flared on gas compressor stations. Therefore, shortage of raw materials for hydrogen production will be.

Catalytic steam reforming reaction of methane carried out at temperatures 750-850 °C.

$$CH_4 + 2H_2O = CO_2 + 4H_2 – 165.3 \text{ kJ } (-39.5 \text{ kcal}) \quad (3)$$

As a catalysts for steam reforming of methane to hydrogen is used mainly oxides of transition metals. The main researches are underway to improve the stability of the catalyst to carbon deposition.

The production of hydrogen from natural gas can significantly improve the economic efficiency of the process [9,10]. It should be emphasized the high added value of the resulting hydrogen product. The price of 1 m^3 of hydrogen is 13,000 tenge, while the price of 1 m^3 of raw materials - natural gas is 180 tenge. Moreover, Kazakhstan has large reserves of natural gas, the country has large reserves of concentrated methane in the gas fields of Kumkol, Karachaganak, as part of natural and associated gas of oil, as well as in coal seams of the Karaganda basin. However, currently methane is used mainly in the form of fuel (domestic and automotive) or is burned in flares at gas compressor stations. Therefore, the production of hydrogen from methane is an economically beneficial and urgent task for Kazakhstan.

2.4 Hydrogen production by photocatalysis

Solar energy is one of the most common sources of energy on Earth, and its inexhaustible source it makes object of research aimed at finding technologies that can efficiently capture, convert and store this energy. Among these technologies photocatalytic hydrogen generation poses a potential alternative to this process [11]. In 1972, when Fujishima and Honda discovered that water can be separated into hydrogen and oxygen on a titanium dioxide (TiO_2) electrode irradiated with ultraviolet light (UV) [12], Photocatalytic hydrogen production has become a promising solution for fuel and environmental pollution problems.However, to use solar energy for hydrogen production, it is necessary to satisfy at least two requirements: (I) it is necessary to develop materials capable of absorbing in the widest range of solar electromagnetic spectra, and (II) these materials are capable of mainly producing hydrogen. The formation of hydrogen for semiconductor materials are mainly affected by the number of photogenerated electrons with respect to the incident photons. Therefore, any process associated with the consumption or generation of electrons which directly affects the efficiency of the low hydrogen [13]. There are three main reasons for such low efficiency with respect to hydrogen in photocatalytic processes: (I) reverse reaction between H_2 and O_2; (II) photogenerated electron-gap recombination(e^- -h^+) and (III) limited light absorption ability.

The catalysts for the photocatalysis of water at this stage are complex, multicomponent oxide semiconductors. These substances are stable during operation, do not lose their original catalytic properties and are not saturated. The effectiveness of famous photocatalysts are not high, but it can be improved by increasing the working surface [14].

The authors of [15] presented the results of a study for the production of hydrogen from water-methanol mixtures on a series of Pd/TiO_2-WO_3 and Pd/P25 catalysts containing a very low amount of Pd (about 0.01 wt.%) are obtained by photodeposition. The introduction of Pd into all carriers led to a significant increase in the rate of hydrogen evolution, despite the small amount of photo-precipitated Pd. Researchers note that, the amount of hydrogen generated increases with increasing methanol concentration.

However, the use of precious metals as an active phase is not economically viable.

The authors of [16] obtained carbon-free $SrTiO_3$ 1D photocatalyst based on polyacrylonitrile (PAN) which synthesized by electrospinning for photocatalytic hydrogen evolution from a water-methanol mixture. The researchers used a combination of solid-state and chemical solution methods andaluminum foil with specific form as a collector for the formation of aligned and oriented $SrTiO_3$/PAN photocatalytic fibers, which will reduce the cost of $SrTiO_3$ synthesis.

Thus, from the literature data suggest that the production of hydrogen photocatalysis has a number of features which should be studied more extensively.

2.5 Conversion of bioethanol

An alternative way to produce hydrogen is the conversion of bioethanol [17]. Bioethanol from biomass is a renewable and cheap starting material for hydrogen production. Bioethanol has several advantages: a) it is available, cheap and is a renewable energy source, b) it is not toxic in contrast to methanol, c) unlike natural hydrocarbons (gasoline, etc.) ethanol does not contain impurities of sulfur-containing compounds that can poison catalysts, used in the reforming of ethanol

The most economically and environmentally efficient is to get the main environmentally friendly energy carrier - hydrogen from bioethanol, because bioethanol from biomass is cheap and renewable raw materials.

In the development strategy on a global scale several directions of the conversion of bioethanol to high-value products dominate [18, 19]: the synthesis of aromatic hydrocarbons, the synthesis of ethylene, the production of hydrogen, the synthesis of acetaldehyde, synthesis-gas synthesis and divinyl synthesis by the Lebedev-Ipatiev method (Fig. 1).

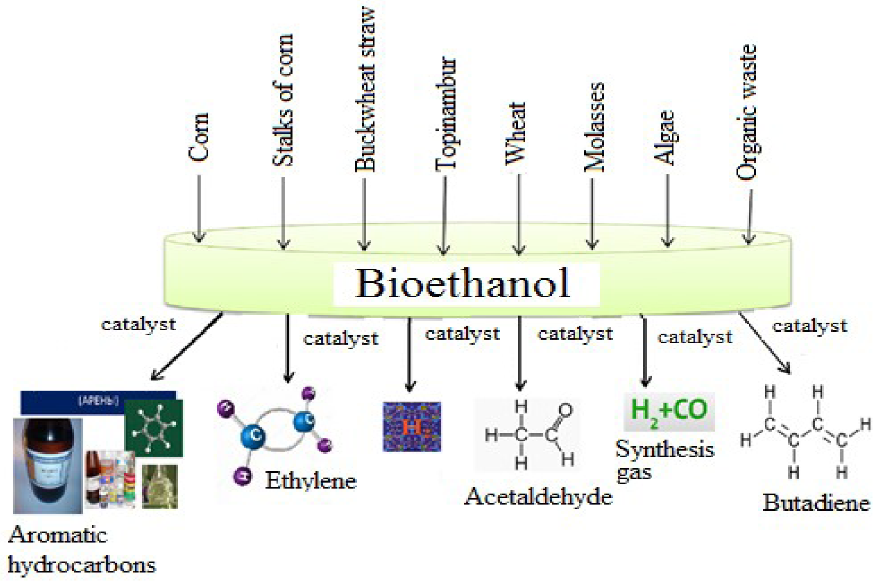

Fig. 1. Raw materials for bioethanol production and its catalytic conversion to valuable products.

One of the strengths of the work is the use of domestic bioethanol as a feedstock. In our country there are plants for the production of bioethanol such as LLP «BM», the capacity of the enterprise is 30 thousand tons of products per year (Zhambyl region); «Biochim» (Taiynshinsky district, North-Kazakhstan region) should resume its work in the first half of 2018. At the construction stage two more factories belonging to LLP «BM» in Taraz and Uralsk. Therefore, there will be no shortage of raw materials, as bioethanol produced in our country will be used as feedstock, in addition, the cost of hydrogen produced from bioethanol is several times higher than the cost of the raw materials – bioethanol.

The special properties of hydrogen (the easiest, having the greatest heat of combustion, etc.) open up a tempting prospect of its application for environmentally friendly energy production. The «hydrogen problem» is now attracting a lot of attention of scientists in the field of science all over the world for many reasons: hydrogen as fuel is economically effective. Industrial interest lies in the fact that the technology of hydrogen production from renewable raw materials of bioethanol is ecologically impeccable. In addition H_2 allows to accumulate large energy reserves, pumping hydrogen to the place of incineration and obtaining energy is 10-15 times cheaper than transporting electricity [20-23]. To celebrate the «hydrogen idea»a large amount of H_2 is needed. One of the possible ways of obtaining such an amount of hydrogen is the conversion of renewable bioethanol feedstock. Development of the technology of hydrogen production from bioethanol in our Republic is not yet developed, although Kazakhstan has huge land. The world community is showing interest in Kazakhstan not only as an oil country, but as a country with rich land resources where there are renewable sources of raw materials.

The creation of a new environmentally friendly technology for the conversion of bioethanol to hydrogen and the design of directional catalysts has not been thoroughly studied. Therefore, the obtaining of scientifically grounded results can positively affect the development of science and technology envisaged in the development strategy of the fuel and energy complex of the Republic of Kazakhstan. The expected social-economic and environmental effect from the implementation of this work is associated with the creation of modern technologies for the production of the main energy carrier H_2 using cheap and renewable bioethanol which meets ecological requirements. The use of cheap domestic bioethanol will provide a significant economic effect. So the cost of the produced hydrogen is 13 000 tenge per 1 m^3, while the price of 1 l of bioethanol is 250 tenge. In addition one of the advantages of using bioethanol is the absence of sulfur-containing and nitrogen-containing toxic compounds in the composition of alcohol conversion products [24-26].

The main processes for producing hydrogen from ethanol are steam reforming (1), partial oxidation (2) and oxidation-steam reforming (3).

$$C_2H_5OH + 3H_2O \rightarrow 2CO_2 + 6H_2 \quad (1)$$

$$C_2H_5OH + 1.5O_2 \rightarrow 2CO_2 + 3H_2 \quad (2)$$

$$C_2H_5OH+(3-2x)\ H_2O+xO_2 \rightarrow 2CO_2+(6-2x)H_20<x<0.5 \quad (3)$$

Ethanol steam reforming (SR) is highly endothermic, which requires the release of large amounts of energy, which increases operating costs. This serious drawback greatly complicates the practical production of hydrogen. The partial oxidation (PO) ethanol is an alternative process for producing hydrogen. Compared from the SR of ethanol, PO is exothermic, and also the advantage of PO is a lower reaction temperature and less coke formation due to the addition of oxygen compared from ethanol steam reforming. However, excessive oxidation can be lead to a lower yield of hydrogen compared from ethanol steam reforming process. Therefore, the integration of processes ethanol SR and PO are represents a reasonable compromise between energy efficiency and yield of hydrogen.Oxidation-steam reforming of ethanol not only provides a favorable energy balance, but also leads to a sufficiently high yield of hydrogen. In addition, the presence of oxygen also contributes to the effective removal of carbon deposits formed during the reaction [27].

Technological conditions of the reaction (reaction temperature, composition of raw materials, space velocity, etc.) and types of catalyst are important control parameters for producing hydrogen from ethanol oxidation by steam reforming. Catalysts are decisive role in terms of the complete conversion of ethanol and maximizing the yield of hydrogen.

Over the last decade, most of the works dealing reformed ethanol mainly focused on the development of active catalysts with a high yield of hydrogen and stability [28-30].

Conclusion

Deep study combining mechanistic reaction analysis and catalyst deactivation, practically not considered, probably due to the complexity of the reaction and its strong dependence on the studied catalysts. Therefore, the establishment of the relationship of the physicochemical characteristics of catalysts with their activity and stability to carbonization is very important.

Thus, a fundamental understanding of the above parameters makes it possible to develop a highly efficient catalyst for the production of hydrogen from renewable bioethanol feedstocks. The conversion of bioethanol to H_2 represents economic, industrial interest and increased social demand, as bioethanol is an affordable, cheap, environmentally friendly and renewable feedstock. In addition, one of the advantages of using bioethanol is the absence of sulfur- and nitrogen-containing toxic compounds in the composition of alcohol conversion products. The creation of a new environmentally friendly technology for the conversion of bioethanol to hydrogen and the design of carbon-stable catalysts have not been thoroughly studied. So, research in this area is one of the priorities in catalysis.

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Перспективные способы получения водорода

Г.Е. Ергазиева[1,3], М.М. Тельбаева[1,2], К. Досумов[1], А.И. Ниязбаева[3]

[1]Институт проблем горения, ул. Богенбай батыра 172, Алматы, Казахстан
[2]Казахский национальный женский педагогический университет, ул. Айтекеби 99, Алматы, Казахстан
[3]КазНУ им.аль-Фараби, пр. Аль-Фараби 71, Алматы, Казахстан

АННОТАЦИЯ

Производство водорода является одним из наиболее перспективных путей развития энергетики будущего. Водород не существует в природе в его элементарной форме и, следовательно, должен быть получен из углеводорода, воды или любых других водородсодержащих соединений. Разнообразие потенциальных источников сырья для получения водорода одна из важных причин, по которой водород является таким многообещающим энергоносителем. В статье рассмотрены способы получения основного энергетического носителя-водорода из природного газа, биоэтанола и др. Среди различных видов сырья биоэтанол очень привлекателен из-за его относительно высокого содержания водорода, доступности, а также безопасности при хранении и обращении.

Ключевые слова: Водород, метан, биоэтанол, катализатор, конверсия, сырье, природный газ, зеленая технология

Сутегіні алудың перспективті әдістері

Г.Е. Ерғазиева[1,3], М.М. Тельбаева[1,2], К. Досумов[1], А.И. Ниязбаева[3]

[1]Жану проблемалары институты, Бөгенбай батыр к-сі 172, Алматы, Қазақстан
[2]Қазақ ұлтты қыздар педагогикалық университеті, Әйтеке би к-сі 99, Алматы, Қазақстан
[3]әл-Фараби атындағы ҚазҰУ, әл-Фараби д-лы 71, Алматы, Қазақстан

АҢДАТПА

Сутегіні өндіру-энергетиканың болашағының дамуының ең перспективті жолдарының бірі болып табылады. Табиғатта сутегі элементті түрде болмайды, сәйкесінше, көміртегіден, судан немесе кез-келген құрамында сутегі бар қосылыстардан алынуы тиіс. Сутегіні алудың патенциалды шикізат көзінің әртүрлілігі - сутегінің көп үміт күтерлік энергия тасымалдаушы болуының маңызды себептердің бірі болып келеді. Мақалада негізгі энергия тасымалдаушы – сутегіні табиғи газдан, биоэтанолдан және т.б. алудың әдістері қарастырылған. Әртүрлі шикізаттардың ішінен биоэтанол құрамындағы салыстырмалы жоғары сутегісімен, қол жетімділігімен, сонымен қатар, сақтау және қолдану қауіпсіздігіне байланысты өте тиімді болып келеді.

Түйінді сөздер: Сутегі, метан, биоэтанол, катализатор, конверсия, шикізат, табиғи газ.