

LOW TEMPERATURE PLASMA AND PLASMA TECHNOLOGIES PLASMA CHEMISTRY FOR CONCEPT OF SUSTAINABLE DEVELOPMENT

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This work is devoted to the exploration of the compatibility of the hybrid plasma-catalytic conversion of liquid hydrocarbons into syngas with the concept of sustainable development. The results of the experimental investigations indicate the high efficiency of plasma-catalytic conversion of ethanol to syngas and the small amount of waste (a few percent of feedstock weight). The results of the simulation of the kinetics using ZDPlasKin code for traditional thermochemical and hybrid plasma-catalytic conversions indicate some differences in their mechanisms, which lead to the significant changes in the syngas ratio.

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INTRODUCTION

The scale on which modern industry uses materials and energy not only led to the substantial depletion of fossil resources but also caused the accumulation of the big amount of wastes, which are hazardous to the environment. In 1980 IUCN (International Union for Conservation of Nature), UNEP (United Nations Environment Program) and WWF (World Wildlife Fund) addressed this problem by developing the World Conservation Strategy for Sustainable Development [1]. In September of 2015, the UN proposed 17 goals of sustainable development, which were adopted by 70 countries [2]. Among these 17 goals, 9 can be achieved with the help of plasma chemistry: "zero hunger", "good health and well-being", "clean water and sanitation", "affordable and clean energy", "decent work and economic growth", "sustainable cities and communities", "responsible consumption and production", "life below water", "life on land".

Among the goals of sustainable development, the minimization of the use of fossil fuels and their replacement with the renewable sources of energy is one of the most important for ecology, society and economics. The switch to the renewable biomass can lead to an ecologically beneficial reduction in the emission of carbon, chemicals and liquid fuels. However, it is widely recognised that the use of first-generation biomass raw materials, such as corn or edible oilseeds, is not a sustainable option in the long run because it competes directly or indirectly with the production of food. Therefore, the European Union is aimed at the use of second-generation biomass raw materials, which includes lignocellulose, waste oils and fats [3]. One of the prioritised ways of achieving the objective of affordable and clean energy is the processing of lignocellulosic biomass into biofuels, but the pace of development of this area is very slow [4]. A major barrier to increasing the scale of biofuels production lies in the disadvantages of two main ways of processing lignocellulosic biomass: thermochemical and biotechnological.

The biotechnical method of biofuel production is based on using the microorganisms for the processing of biomass into the desired product. This technology is

used successfully to obtain ethanol from plant sugars and starches, but it faces significant challenges in the transition to the more complex raw materials, such as lignin and cellulose, and while attempting to produce more sophisticated biofuels than ethanol [5]. Biological agents used for the processing of raw materials are selective to the composition of these raw materials, sensitive to process conditions, and their reuse is complicated. Complex raw materials need to be additionally pretreated by the toxic substances, such as acids and alkalis, for the biotechnical processing. This treatment leads to the formation of toxic wastes in the raw materials, which poison both the microorganisms and the environment. At this stage of technology development, the performance of the biotechnical processing of lignocellulosic feedstock is low and is not able to provide the required rate of biofuel production growth.

The modern thermochemical technology of lignocellulosic biomass processing requires much higher temperatures than biotechnological methods (over 700°C for pyrolysis and 1000°C for gasification). A significant disadvantage of the thermochemical technologies is low energy efficiency. It is also worth noting that the use of high temperatures not only complicates the design of processing devices and requires the increase of safety requirements, but also decreases the selectivity of the raw materials conversion and leads to the emergence of byproducts. Therefore, the traditional thermochemical (temperature > 700°C) and traditional plasma-chemical (temperature > 1000°C) (which is based on the ideas of thermochemical conversion using plasma torches) processing of biomass leads to the appearance of the unwanted byproducts and wastes. In the case of catalytic conversion, a high content of impurities in the initial raw biomass negatively affects the yield and longevity of catalysis.

As of today, the discussion is open on the development of principles behind the construction of new technologies for the conversion of substances, which are known as "green chemistry", which is a departure from the traditional evaluation of the effectiveness using the chemical yield to the assessment of the cost-effectiveness based on the lack of hazardous

waste and non-toxic and/or hazardous substances. "Green chemistry" has to transform the raw materials (preferably renewable), exclude hazardous waste, and use no toxic agents in the production of chemical products and in their application [6]. There should be the development of processes involving H₂, O₂, CO, CO₂, NH₃ as a direct source of atoms H, O, C and N in the production of fine chemicals. The quantitative measure of the environmental acceptability of chemical technology is E factor, which is defined as the ratio of waste weight to the weight of a target product. Waste includes everything that is not a target product [6].

Developed over the past decade chemo-catalytic and/or biocatalytic integration technologies of the "green chemistry" have not yet led to the large-scale processing of lignocellulosic biomass into synthesis gas, and do not fully meet the requirements of sustainable development [5].

In recent years, very promising results have been obtained in the study of the reforming of hydrocarbons into synthesis gas in plasma-catalytic systems. These systems are the improvement over the thermochemical systems and have gas-discharge plasma used additionally as a catalyst. The main result, which gives reasons to consider plasma-catalytic approach as a promising new alternative to traditional conversion technologies with regards to the Arrhenius kinetics of chemical reactions, is the reduction of the temperature of the reforming process by several times.

The plasma-catalytic approach uses active particles (OH, O, H, A_i⁺, A_j⁻, e, A_k^{*e,v,f}, hv), which are generated from the non-toxic and/or safe reagents in the plasma to initiate the chain reactions of renewable biomass conversion at low temperatures (~ 250...350°C). Renewable biomass can be converted directly into hydrocarbons (lower olefins, alkanes and aromatic substances), which form the basis of petrochemical plants raw materials, or into synthesis gas, which can be converted into liquid fuel or chemicals [7] using established technology (Fischer-Tropsch process, methanol synthesis).

This work is devoted to several topics: further experimental research of hybrid plasma-catalytic system with the gas discharge generator of the wide-aperture flow of non-isothermal plasma, the investigation of some features of the kinetics in a plasma of wide-aperture rotating gliding discharge using numerical modelling, the comparison of chemical kinetics during thermochemical reforming and reforming stimulated by the additional injection of radicals that work as the seeds of the chain chemical processes of reforming.

1. METHODS AND EXPERIMENTAL SETUP

Partial oxidation reforming of hydrocarbons is the most energy-efficient and economical reaction pathway for use in the conversion methods that utilise electric energy. This is especially important for the plasma-chemical reforming methods that use electric energy. The reason for this is that in modern power plants the production of one unit of electrical energy requires three units of chemical energy. Fig. 1 shows the schemes of two hydrocarbon conversion approaches are present in

plasma-chemistry: plasma reforming and plasma-catalytic reforming.

In plasma approach, the hydrocarbon and oxidant are introduced directly into the discharge area after which the activated mixture is injected into the reaction chamber. Such approach leads to the appearance of the exothermal reactions in the discharge area, which increase the rotational temperature of heavy plasma components and cause the decrease of plasma non-isothermality and drop in the yield of desired products [8,9]. In plasma-catalytic approach, only a part of the oxidant is introduced into the discharge area and the activated oxidant is injected in a form of radicals into the reaction chamber, which has the separate injection of hydrocarbon. This approach results in the generation of the non-isothermal plasma, which provides the increased selectivity of desired product. Discharge can be used to activate a part of the oxidant while the rest of the oxidant is introduced into the reaction chamber together with the hydrocarbon.

A discharge chamber, which is attached to the reaction chamber [10-13], is designed for the plasma-catalytic approach to the hydrocarbon conversion. Plasma is generated using wide-aperture rotating gliding discharge [14-18]. The discharge is powered using DC power source. Fig. 2 shows the discharge chamber, gas flow directions and wide-aperture rotating gliding discharge, which was used for the plasma-catalytic conversion of hydrocarbons.

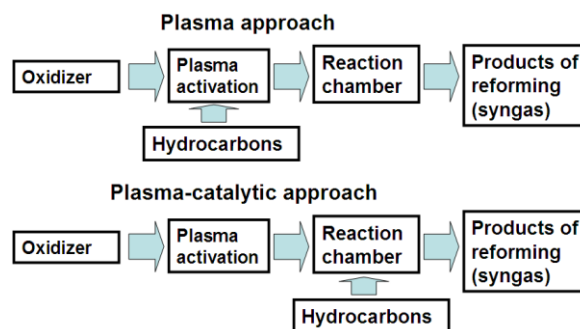


Fig. 1. Schematic representation of plasma and plasma-catalytic approaches to hydrocarbon conversion

96 % ethanol was used as a model hydrocarbon during the plasma-catalytic reforming of hydrocarbons, atmospheric air, which was supplied using a compressor, was used as an oxidant. The system for plasma-catalytic reforming of hydrocarbons into the synthesis gas was designed for the output power of 25 kW and tested using 53.3 g/min (25 kW) ethanol flow. The experimental laboratory studies of hybrid plasma-catalytic reforming of ethanol were conducted at lower output power. Optical emission spectroscopy was used for the study of plasma. Gas chromatography was used for the study of gas-phase products. Numerical modeling was used for the study of kinetics of plasma and thermochemical reforming.

2. EXPERIMENTAL RESULTS

The hybrid plasma-catalytic approach can become a new alternative to the traditional conversion technologies. The plasma-catalytic approach uses the

active species (OH, H, O), which are produced inside plasma from the non-toxic and/or safe reagents, to initiate the chain reactions of renewable biomass conversion at a low temperature (~ 250...350°C).

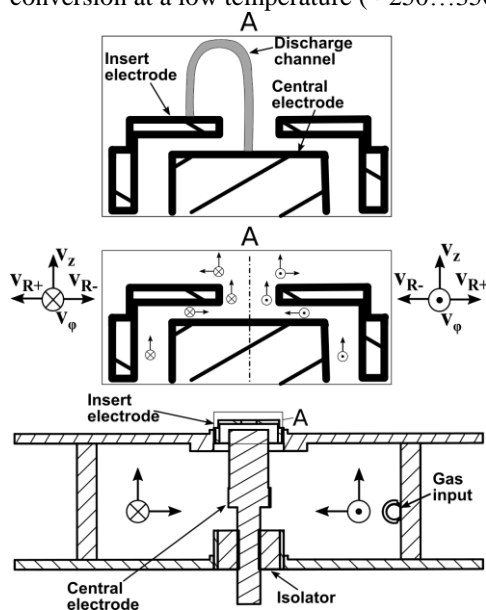


Fig. 2. Scheme of discharge chamber, directions of gas flows and wide-aperture rotating gliding discharge during plasma-catalytic reforming of hydrocarbons

Fig. 3 shows the dependence of hydrogen energy yield on the reaction chamber temperature during the plasma-catalytic reforming of ethanol. Discharge power is 24 W, discharge current is 60 mA, air flow is 7 l/min (the amount of air is 40 % higher than required by the stoichiometry of ethanol partial oxidation reaction), ethanol flow is 4.3 g/min (2 kW). Discharge activated only 20 % of the total amount of air. Maximum hydrogen (desired product) energy yield was reached at

the temperature of 250°C, which is significantly lower than for the traditional thermochemical (> 700°C) and plasma-chemical (> 1000°C) hydrocarbon conversion.

Fig. 4 shows the comparison between the hydrogen energy yields and conversion efficiency of the hybrid plasma-catalytic reforming of ethanol (the best results) and other methods of hydrogen production that utilise electrical energy. Comparison of hydrogen production methods includes both commercial methods, such as water electrolysis [19] and biomass gasifiers based on the Westinghouse/AlterNRG plasma torches [20], and results of the most advanced studies on the production of hydrogen from ethanol using low temperature atmospheric pressure plasma, such as Laval nozzle arc [21], dielectric barrier discharge [22], MW discharge [23], arc discharge [24], GEN3 plasma source [25], submerged nanosecond pulsed discharge in the water solution of ethanol [26].

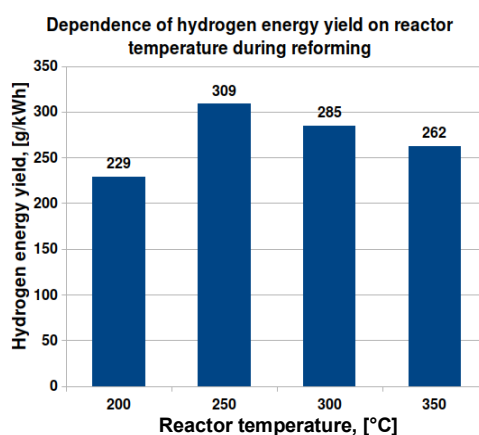


Fig. 3. Dependence of hydrogen energy yield on reaction chamber temperature

Hydrogen energy yield and reforming efficiency in systems that use electricity for hydrogen production

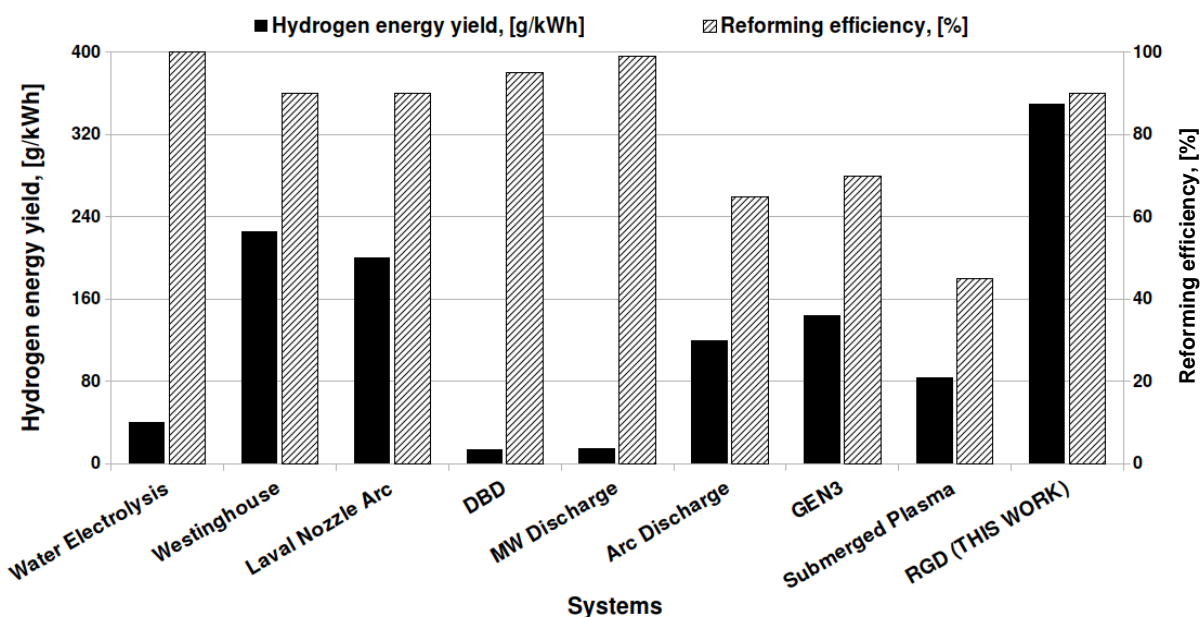


Fig. 4. Comparison of the hydrogen energy yield and conversion efficiency of hybrid plasma-catalytic reforming of ethanol (RGD (THIS WORK)) with other methods of hydrogen production using electrical energy: Water Electrolysis [19], Westinghouse [20], Laval Nozzle Arc [21], DBD [22], MW Discharge [23], Arc Discharge [24], GEN3 [25], Submerged Plasma [26]

The hydrogen energy yield of hybrid plasma-catalytic ethanol reforming is the highest in comparison with the other methods [19-26]. The ratio of the reactor power output to its volume is approximately 100 kW/l. The hybrid plasma-catalytic approach can be used to solve the existing problems of the traditional renewable biomass reforming methods.

3. RESULTS OF NUMERICAL MODELING

The kinetics of plasma-chemical processes in the plasma of wide-aperture rotating gliding discharge and chemical kinetics of ethanol conversion into synthesis gas were calculated via ZDPlasKin programme code [27, 28]. ZDPlasKin allows the simulation of plasma kinetics by taking into account the preliminary set of input parameters of the studied system (temperature, pressure, the initial concentration of reagents etc.). During the calculation fulfilled for the plasma of moist air, the database used for the calculation comprised of 900 chemical and electron-molecular reactions involving 83 components. Time sampling step was 10^{-12} s. The time interval for the calculation of changes in the concentration of plasma components at the time evolution was 10^{-3} s. It is significantly higher than the time of the exposure of gas to the current channel of gas discharge considering the experimental data: the rate of gas flow was $\sim 10^3$ cm/s, wide-aperture rotating discharge is a transverse discharge with the diameter of current channel $\sim 10^{-1}$ cm, respectively the time of gas exposure to the current channel is $\sim 10^{-4}$ s.

This list of reactions is based on a mechanism proposed in [29]. Because the experimental data confirms that the rotational temperature is high enough (~ 2500 K), all the reaction of the oxidation of atmospheric nitrogen were included to the list of reactions. The mechanism was complemented by the reactions, which involved such species as N_2O , NO_2 , NO_3 , N_2O_5 , N_2O^+ , NO_2^+ , N_2O^- , NO_2^- , NO_3^- , the rates of chemical and electron reactions were taken from [30]. According to the experiment, the electric field in the plasma zone varied in the range of 100...500 V/cm, which necessitated the addition of dissociation reactions by electron impact of such components as H_2O , NO_2 and HO . At the same time, in this type of conversion, the air with the addition of H_2O is the plasma gas, which is why the list of reactions involving H_2O and its derivatives was added to this mechanism. The list of reactions involving H_2O was composed using [31, 32].

Typical time dependencies of the concentrations of N_2 , N , O_2 , O , O_3 , H , OH , H_2 , which are calculated for the plasma of gas discharge – $[A_i](t)$, are shown in Fig. 5.

Provided dependences $[A_i](t)$ show that during the experiment the setting time of the fixed values $[A_i]$ of O , H , and OH radicals corresponds to $\sim 10^{-4}$ s, and the degree of the dissociation of molecular oxygen O_2 and water H_2O is $\sim 10^{-2}$.

During the calculation of chemical kinetics in the reactor zone of the experimental system [10], the database contained 130 chemical reactions involving 30 component [33]. Time sampling step was 10^{-6} s. The time interval for the calculation of changes in the concentration of plasma components was 10^{-3} s. The

time interval for the calculation of changes in the concentration of the gas components at the outlet of the reactor zone (Fig. 6) was equal to 10 seconds.

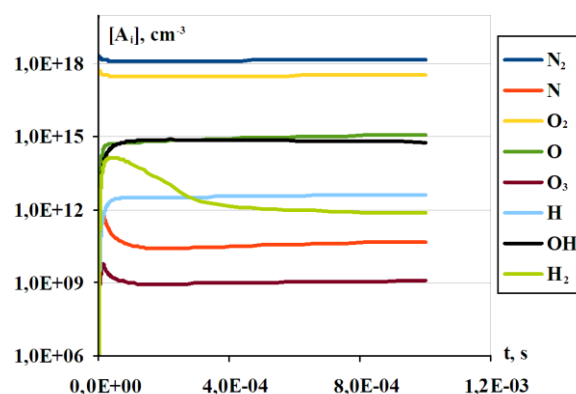


Fig. 5. Time dependence of concentrations of N_2 , N , O_2 , O , O_3 , H , OH , H_2 in plasma of gas discharge in air at atmospheric pressure, 10% humidity, 2500 K gas temperature, and at 400 V/cm electric field

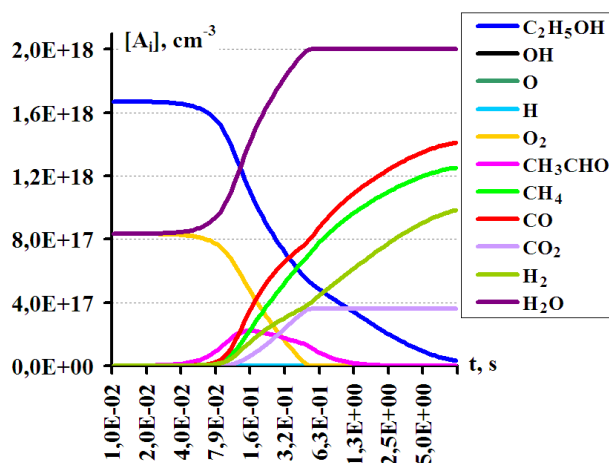


Fig. 6. Time dependence of C_2H_5OH , OH , O , H , O_2 , CH_3CHO , CH_4 , CO , CO_2 , H_2 , H_2O concentrations for the thermochemical conversion of ethanol at 1100 K for $1C_2H_5OH/0.5O_2/0.5H_2O/2N_2$ initial mixture

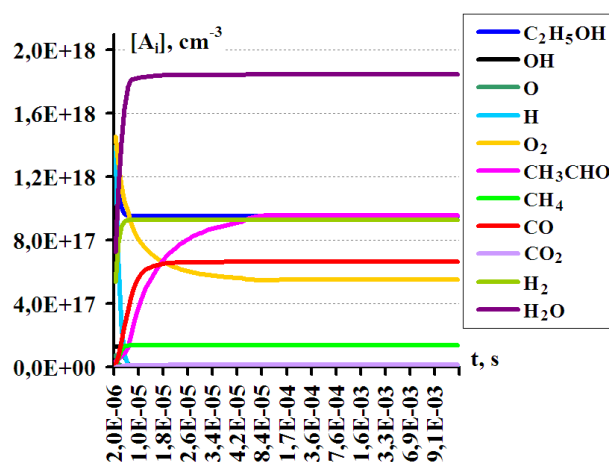


Fig. 7. Time dependence of C_2H_5OH , OH , O , H , O_2 , CH_3CHO , CH_4 , CO , CO_2 , H_2 , H_2O concentrations for the thermochemical conversion of ethanol at 523 K for $1C_2H_5OH/1O/0.5H/0.5OH/2N_2$ initial mixture

It is significantly higher than the time of the exposure of gas to the reactor zone with regard to the experimental data: the rate of gas flow is $\sim 10^3$ cm/s, typical reactor size is approximately 10 cm, thus the time of the exposure of gas to the reactor zone is $\sim 10^{-2}$ s.

The typical calculated time dependences of C_2H_5OH , OH , O , H , O_2 , CH_3CHO , CH_4 , CO , CO_2 , H_2 , H_2O concentrations for the thermochemical conversion of ethanol at 1100 K for $1C_2H_5OH/0.5O_2/0.5H_2O/2N_2$ initial mixture and at 523 K for $1C_2H_5OH/1O/0.5H/0.5OH/2N_2$ initial mixture are shown in Fig. 6 and Fig. 7 respectively.

The comparison between the time dependences of concentrations, which are shown in Fig. 6 and Fig. 7, allows for the determination of the effect caused by the stimulation of the thermochemical conversion using O , H and OH radicals, which are the seeds of the chain conversion process, injected from plasma into the reactor zone. The comparison of the gas composition calculated under conditions of full thermochemical conversion of ethanol (see Fig. 6) at the outlet of the reactor zone and reforming stimulated by the additional injection of radicals (see Fig. 7) also shows the existence of difference in the conversion mechanism, which results in significant changes in the syngas ratio $[H_2]/[CO]$.

CONCLUSIONS

The experimental and theoretical studies of plasma-catalytic and purely thermochemical conversions of ethanol show that:

1. Plasma-catalytic approach is an emerging alternative to the traditional conversion technologies. Plasma-catalytic approach uses active species (OH , O , H) generated in the plasma from non-toxic and/or non-hazardous reagents to initiate the chain reactions of renewable biomass conversion at low temperature ($\sim 500 \dots 600$ K).

2. Plasma-catalytic approach confirms the concept of sustainable development with the production of small amounts of waste ($\sim n$ %).

3. Studies of chemical kinetics revealed differences in the mechanism of the conversions that result in significant changes in syngas ratio ($[H_2]/[CO]$).

ACKNOWLEDGEMENTS

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ПЛАЗМОХИМИЯ ДЛЯ КОНЦЕПЦИИ УСТОЙЧИВОГО РАЗВИТИЯ

В.Я. Черняк, О.А. Недыбалиук, А.Н. Цымбалиук, И.И. Федирчик, Е.И. Чунихина, Е.В. Мартиш, В.В. Юхименко, Ю.П. Веремий, И.В. Присяжневич, Е.В. Присяжная

Работа посвящена изучению совместимости гибридной плазменно-каталитической конверсии жидких углеводородов в сингаз с концепцией устойчивого развития. Результаты выполненных экспериментальных исследований указывают на высокую эффективность плазменно-каталитической конверсии этанола в сингаз с малым количеством отходов (несколько процентов от массы исходного сырья). Результаты проведенной симуляции кинетики с использованием программного кода ZDPlasKin традиционной термохимической и гибридной плазменно-каталитической конверсий указывают на некоторые отличия в их механизмах, которые приводят к существенному изменению сингазного соотношения.

ПЛАЗМОХІМІЯ ДЛЯ КОНЦЕПЦІЇ СТАЛОГО РОЗВИТКУ

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Робота присвячена вивченню сумісності гібридної плазмово-каталітичної конверсії рідких вуглеводів у сингаз із концепцією сталого розвитку. Результати виконаних експериментальних досліджень вказують на високу ефективність плазмово-каталітичної конверсії етанолу в сингаз з малою кількістю відходів (декілька відсотків від маси вихідної сировини). Результати проведеної симуляції кінетики з використанням програмного коду ZDPlasKin традиційної термохімічної та гібридної плазмово-каталітичної конверсії вказують на деякі відмінності їхніх механізмів, які спричиняють істотні зміни сингазного співвідношення.