

## CHEMISTRY

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### Review of Some Methods of CO<sub>2</sub> Utilization

#### Abstract

Protecting the environment from harmful substances (carbon oxides, nitrogen oxides, soot products, etc.) is one of the pressing issues of our time. This article discusses specific topics related to the need to reduce carbon dioxide emissions into the atmosphere, which are generated during biomass gasification, Fischer-Tropsch synthesis, and other processes. The article also identifies alternative methods for producing valuable petrochemical products, which significantly reduce gas emissions into the atmosphere.

**Keywords:** oil, gas, coal, peat, carbon dioxide

#### Introduction

One of the important directions of scientific and technological development is the creation and implementation of new technologies, substances, and materials that ensure resource conservation and meet environmental requirements. In the general concept of "sustainable development of civilization" adopted by the UN, the primary focus is on developing environmentally safe technologies that prevent the release of harmful substances into the atmosphere, utilize existing industrial waste, use non-renewable natural resources rationally, and recycle materials after their service life ends (Gasimova, 2023, p. 153; Kakhramanov, 2022; Gasimova, 2023; Ismayilova, 2024, pp. 25-26).

#### Research

The extensive use of oil, gas, coal, peat, and other fossil fuels in energy production and manufacturing has led to a deterioration in environmental indicators, particularly the accumulation of greenhouse gases in the atmosphere. It has been calculated that over the last 250 years, carbon dioxide concentration has increased by one-third, and methane by one and a half times (Kreinin, 2010, p. 396; Kalekin, 2012; Kadzhiyev, 2011). The increased use of diesel vehicles has contributed to a rise in the share of nitrogen oxides and soot in exhaust gases.

In 1997, the Kyoto Protocol on reducing greenhouse gas emissions was signed in Kyoto, Japan, to curb global warming. Greenhouse gases (CO<sub>2</sub>, methane) absorb infrared rays, potentially leading to an increase in air temperature, glacier melting, and a rise in sea levels (Ilinsky, 2003, pp. 25-26). Forests partially absorb CO<sub>2</sub>; however, the amount of carbon dioxide released into the atmosphere has become so large that the "green lungs of the planet" can no longer process it entirely. The main source of CO<sub>2</sub> emissions into the atmosphere is flue gases from various types of energy installations. A single large thermal power plant (TPP) operating on natural gas consumes between 0.5 to 2 million cubic meters of gas per day, leading to CO<sub>2</sub> emissions of 1,000 to 4,000 tons per day. If carbon dioxide in such quantities were used as a raw material for the production of

hydrocarbons or other petrochemical synthesis products, it would result in a large petrochemical complex producing at least 2 million tons of products per year.

The Kyoto Protocol, which regulates greenhouse gas emission limits, has been extended until 2020. About 200 countries, including 38 industrially developed ones, supported the extension. However, the United States, China, India, Japan, Russia, and Canada (which officially withdrew from the agreement) do not intend to participate in the new commitment period. Therefore, entirely new approaches are needed for environmental protection, as no agreement can stop the accumulation of harmful substances in nature unless specific measures are taken to improve existing technologies and implement more progressive ones (Juang, 2010, pp. 3346-3364; Zasa, 2011).

Let's consider recent developments on carbon dioxide utilization and the development of new methods for integrating it into technological processes for synthesizing important petrochemical products (Patent, No. 2349371, 9304247, 2458005, 2385836, 2036900, 2455048; White, 2004, pp. 4-10).

One method proposed (Patent No. 2478074, 2001) involves the complete utilization of CO<sub>2</sub> produced during underground coal gasification (UCG). In this method, coal gasification is carried out in situ using a series of blast and gas extraction wells in an underground gas generator, and CO<sub>2</sub> is captured from the burning gas mixture in a surface chemical complex. The CO<sub>2</sub> captured in the surface chemical complex is divided into two streams. The first stream is injected into the blast wells of the operating underground gas generator, initiating the endothermic reaction according to the scheme:  $\text{CO}_2 + \text{C} \rightarrow 2\text{CO} - 173 \text{ kJ/mol}$ .

This enriches the UCG with carbon monoxide. The second stream of CO<sub>2</sub> is injected into the exhausted gas generator for storage. Injection of CO<sub>2</sub> is stopped when its concentration (in periodically taken samples) reaches 90 %. The UCG gas undergoes sequential preparation for use in the surface complex: cooling, purification, conversion of CO to CO<sub>2</sub> ( $\text{CO} + \text{H}_2\text{O} \rightarrow \text{CO}_2 + \text{H}_2$ ), H<sub>2</sub>S removal, and finally, CO<sub>2</sub> utilization.

The remaining combustible components (CO<sub>2</sub> + H<sub>2</sub> + CH<sub>4</sub>) are directed to the consumer. This CO<sub>2</sub> utilization method is planned for implementation at an experimental underground gas generator (in Kuzbass).

Known UCG technologies also involve drilling a series of blast and gas extraction wells on a coal seam intended for coal gasification in situ, capturing the gas mixture in a surface chemical complex (Patent No. 2293845, 2007).

A patent (Patent No. 2513947, 2011) describes a method of partial CO<sub>2</sub> utilization based on injecting CO<sub>2</sub> and water into the underground gas generator, allowing for the regulation of UCG gas composition. However, this technology does not fully solve the problem.

In the chemical industry, the so-called "monoethanolamine method" of CO<sub>2</sub> utilization is widely used, based on absorbing CO<sub>2</sub> from gas mixtures according to the reaction:  $\text{HO}(\text{CH}_2)\text{NH}_2 + \text{CO}_2 \rightarrow \text{HO}(\text{CH}_2)\text{NHCO}(\text{OH})$ .

The monoethanolcarbamate obtained by this method is thermally unstable and easily decomposes by the reverse reaction. Therefore, it cannot be used in underground coal gasification.

Technologies have been developed to use carbon dioxide for injection into oil reservoirs to enhance oil recovery. The CO<sub>2</sub> injection method to increase oil recovery is applied in practice.

It should be noted that the requirements for CO<sub>2</sub> injection into hydrocarbon reservoirs to increase oil recovery differ from those for underground storage. In recent years, researchers have focused on not only CO<sub>2</sub> utilization but also its storage. The fact is that the amount of CO<sub>2</sub> needed for reservoir oil recovery constitutes a much smaller part than that injected into underground storage.

A method has been developed for underground CO<sub>2</sub> storage in a porous and permeable hydrocarbon reservoir, which has at least one injection well and at least one production well, and includes the following stages:

1. Extraction of a production fluid stream from the production well, consisting of produced hydrocarbons, water, and CO<sub>2</sub>.

2. Directing the production fluid stream to a processing facility, where a vapor-phase stream containing CO<sub>2</sub> and volatile hydrocarbons is separated.
3. Compressing the vapor-phase stream to a pressure above the maximum at which gas and liquid phases can coexist.
4. Cooling the compressed stream (resulting in a cooled stream in the vapor phase).
5. Directing the incoming CO<sub>2</sub> stream from the feed to the injection equipment (it can be either in liquid phase or supercritical state).
6. Mixing the cooled stream from stage 4 with the incoming CO<sub>2</sub> stream from the feed (forming a single stream).
7. Injecting the combined stream into the hydrocarbon reservoir through the injection well.

The hydrocarbon reservoir represents a hydrocarbon-bearing geological horizon with an underlying aquifer communicating with the hydrocarbon-bearing geological horizon, and the aforementioned combined stream is injected into the aquifer.

The storage of carbon dioxide, which is formed in large quantities in many chemical processes as a by-product, is also an urgent problem, such as in ammonia production, hydrogen production by hydrocarbon feedstock reforming, and others.

Hydrogen and syngas production has become one of the priorities in modern energy and basic organic synthesis in recent years. Therefore, finding rational ways to produce additional hydrogen is important. Among the alternative methods of hydrogen production, steam catalytic conversion (SCC) of hydrocarbons is currently the most widely used industrial process in the world for oil refining and petrochemistry.

SCC primarily uses natural and plant gases, as well as straight-run gasoline. The process occurs in two stages:



where  $n$  and  $m$  are the number of carbon and hydrogen atoms, respectively, in the hydrocarbon molecule. The higher the hydrogen content in the raw material, the higher the hydrogen yield. In this regard, methane is the most favorable raw material (hydrogen content in it is 23 %). The source of methane is, as is known, natural gases (CH<sub>4</sub> concentration is 94-95 %). Cheap dry gases from oil refining can also be used for hydrogen production. SCC is a fairly energy-intensive process; the first stage of the process is highly endothermic. The second stage occurs with heat release (in methane conversion,  $Q=206.7$  kJ/mol), and the process can be carried out without a catalyst at temperatures above 1000°C or in the presence of catalysts at 800-900°C.

In modern processes, special attention is given to the processing of natural, associated, and waste gases, as well as biogas. Fedotov and others consider processes conducted on catalytic membranes prepared from Ni(Al) and Co<sub>3</sub>O<sub>4</sub> powders in a 1:1 ratio to be promising (Fedotov, 2014, pp. 309-311). They discovered a non-additive increase in the catalytic activity of these systems during the dry reforming of methane compared to the sum of the activities of Ni- and Co-containing membranes. It was established that the specific productivity of synthesis gas from the bimetallic sample reaches 85,000 L/h·dm<sup>3</sup>, exceeding similar performance for membranes with other nickel-cobalt component ratios. The dry reforming process is carried out on a membrane-catalytic setup in a flow reactor at temperatures ranging from 400 to 800°C with a CH<sub>4</sub> ratio of 1:1. The feed rate varies from 20 to 750 L/h (Fedotov, 2014, pp. 339-340). The pressure at the membrane inlet is 1.1 atm, and 1 atm at the outlet. The high selectivity of the developed catalytic system is attributed by the authors to the nano-sized Ni-Co alloy particles found in the membrane structure, formed on the surface of  $\gamma$ -Al<sub>2</sub>O<sub>3</sub>.

Nickel-cobalt-containing catalysts were also used in high-selectivity gasification processes of carbon dioxide from biomass fermentation products under the guidance of Academician I.I. Moiseyev (Moiseyev, 2001, pp. 5-10).

Biomass gasification is seen as an important method for linking biomass carbon with products of basic organic synthesis. The hydrocarbon components of biomass are thermally unstable and degrade under relatively mild conditions.

The main thermolysis routes include carbon-releasing reactions:



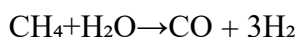
Stoichiometrically, thermolysis with the formation of synthesis gas according to the following scheme is also possible:



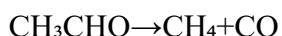
In a study, a two-stage gasification scheme was first proposed, allowing for the production of synthesis gas from any fermentable carbohydrate biomass. In the first stage, the raw material undergoes fermentation:  $\text{C}_6\text{H}_{12}\text{O}_6 \rightarrow 2\text{C}_2\text{H}_5\text{OH} + 2\text{CO}_2$  (3)

In the second stage, the alcohol and the resulting carbon dioxide undergo catalytic reforming to produce synthesis gas:  $\text{C}_2\text{H}_5\text{OH} + \text{CO}_2 \rightarrow 3\text{CO} + 3\text{H}_2$  (4)

During fermentation processes, by-products (such as fusel alcohols, i.e., higher molecular weight alcohols) are also formed. It has been established that these products can be used to obtain energy carriers through carbon dioxide reforming using ceramic nickel-cobalt porous membrane catalysts. Samples of porous ceramic membranes were prepared by the method of self-propagating synthesis. Precursors included Ni metal powders, 5 % Al metal, and mixtures of Ni metal containing aluminum and cobalt oxide ( $\text{Co}_3\text{O}_4$ ). Materials with various Ni and Co contents (with ratios of 19, 4, and 1) were obtained. It was found that complete ethanol conversion (according to reaction 4) on all membranes was achieved at 400°C, while on a granular catalyst of identical composition, methane formation from ethanol was observed at 700°C. Further temperature increases led to a decrease in methane content, likely due to the enhancement of endothermic reactions of its transformations according to the following schemes:



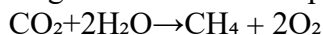
In the temperature range of 300-400°C, the formation of hydrogen, acetaldehyde, and methane was observed:



With an increase in temperature to 420-430°C, the acetaldehyde concentration decreases, with the intensive formation of CO and CH<sub>4</sub>.

Thus, these studies have demonstrated the importance of using effective catalytic systems in biomass thermolysis processes to increase technically important raw material resources. At the same time, it is necessary to consider potential reactions that could lead to CO<sub>2</sub> formation outside of a closed cycle.

To maintain ecological balance, the limiting stage of the global CO<sub>2</sub> cycle should be its formation rather than its consumption. Due to extensive industrialization and human activities, especially in recent years, the dynamics of CO<sub>2</sub> utilization have lagged behind its usage (as building blocks in the synthesis of various organic compounds). Various methods for chemical utilization of CO<sub>2</sub> have been proposed (Rozovsky, 1980). For instance, a process for obtaining CH<sub>4</sub> and O<sub>2</sub> by reacting CO<sub>2</sub> with water vapor in the presence of Ni, Co, or Rh-based catalysts has been developed:



This process takes place at high temperatures in the presence of rare catalysts. Carbon dioxide and ammonia can also be used to synthesize urea. The process is carried out at high pressure (approximately 200 kg/cm<sup>3</sup>) at a temperature 40°C lower than in the main reactor. This technology is complex and requires special equipment.

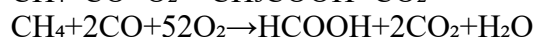
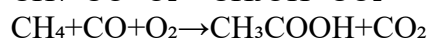
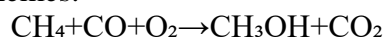
A method for utilizing carbon dioxide in closed systems and industry has also been described. The method involves passing a gas or gas mixture through a substance with the empirical formula  $(a \cdot \text{CaO}) \cdot (b \cdot \text{V}_2\text{O}_5) \cdot (c \cdot \text{M}_2\text{O}) \cdot (d \cdot \text{MO}) \cdot (e \cdot \text{M}_2\text{O}_3) \cdot (f \cdot \text{MO}_2) \cdot (g \cdot \text{M}_2\text{O}_5)$  (where M is a metal, and a, b, c, d, e, f, g are molar fractions, with a as (2-4):1) activated by radiation with a wavelength of 370 nm at 250-800°C. The reaction completeness is 98-100 %. However, this method is not sufficiently effective due to the complex CO<sub>2</sub> utilization system, which includes radiation use and high temperature conditions.

A new method for CO<sub>2</sub> utilization has been proposed, which could find applications in the

chemical industry. This method uses trifluoroacetic acid (TFA) saturated with oxygen (TFA-O<sub>2</sub>). CO<sub>2</sub> is passed through a container filled with TFA-O<sub>2</sub> at a temperature of 10-25°C and atmospheric pressure, forming a resin-like product – a mixture of products with the formula (C<sub>4</sub>H<sub>7</sub>O<sub>9</sub>)<sub>n</sub>, where n ranges from 2 to 9. The breakthrough point of CO<sub>2</sub> through the first container is recorded, after which it is fed into a second container filled with a fresh portion of TFA-O<sub>2</sub>. The spent TFA is sent for regeneration, which involves its saturation with a new portion of oxygen, after which TFA-O<sub>2</sub> is reused in the process.

The authors of this development, Vishnevskaya M. V., Ivanova M. S., and others, propose using the resulting hydrocarbon mixture as an octane-enhancing additive to motor fuel. The use of 1-2 % by mass (of the base gasoline) allows for an increase in octane number by 10-12 points (according to the research method). It is noted that this additive improves fuel stability, reduces losses during storage and transportation, improves fuel cleaning properties, removes deposits in the engine's intake system, and reduces the amount of toxic substances in the exhaust gases. The CO<sub>2</sub> utilization rate is approximately 100 %.

The developed process is simple, carried out at low temperatures and atmospheric pressure. However, TFA used in the oxidation process is a toxic substance. Trifluoroacetic acid was also used by E. G. Chepaikin, A. P. Bezruchenko (2010), and others in the catalytic oxidation reaction of natural gas and associated oil gas to develop single-stage methods for obtaining basic oxygenated products. In oxidative functionalization of C1-C4 alkanes under the influence of O<sub>2</sub> and CO, catalytic systems consisting of Rh, Pd, and Pt halides and copper compounds were used (Chepaikin, 2002). The reaction was carried out in aqueous trifluoroacetic acid according to the following schemes:

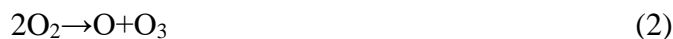


It was found that the oxidation of alkanes proceeds not only through C-H bonds but also through C-C bond cleavage. The problem of direct oxidation of alkanes in the presence of metal complex catalysts is associated with the activation of C-H bonds and molecular oxygen (Chepaikin, 2006). E.P. Chepaikin and others studied the mechanism of action of the Rh- and Pd-containing catalysts they developed in the oxidation reactions of lower alkanes. The authors found that catalytic systems effectively operate in aqueous trifluoroacetic acid. To convert oxygen into an active state, a reductant (carbon monoxide) is introduced, and co-catalysts (iodine, iron, copper compounds) are used. In catalysis with rhodium or palladium complexes, co-catalysts are reduced by carbon monoxide to a low-valent state, in which they can interact with oxygen. Attempts were made to replace the corrosive and toxic trifluoroacetic acid with less toxic solvents (tetrahydrofuran, acetonitrile, or ionic liquids), but replacement was unsuccessful; the activity of the catalysts decreased (Chepaikin, 2014).

The method for utilizing carbon dioxide in an aquifer was developed by A. A. Barenbaum, S.N.Zakirov, and others (Patent No. 2514076, 2014). For this purpose, a subsurface aquifer is selected, preferably one that has an outlet to the Earth's surface and a recharge area in the form of a river, sea, lake, and a generally active filtration regime. At least one local trap is identified, and the selected aquifer and local trap are prepared for industrial use by determining the chemical composition of the water in the aquifer, as well as the composition of the rocks through core samples. The rocks are selected to contain compounds of iron group metals (Fe, Ni, Co, Mo, SiO<sub>2</sub>, Al<sub>2</sub>O<sub>3</sub>), as well as clays and zeolites. These act as catalysts in the polycondensation synthesis of hydrocarbons, as well as hydrogen and oxygen from CO<sub>2</sub> and water. This method can be applied to depleted oil and gas fields to form new deposits (Patent No. 2036900, 2011).

In, processes for the formation of CO<sub>2</sub> and water during the purification of gases from harmful impurities are considered, which involve the decomposition of gases into corresponding products and their subsequent oxidation by oxygen in the high-frequency discharge zone. The decomposition scheme, for example, of polyurethane foam and the subsequent oxidation of the resulting products in the discharge zone can be represented as follows:





Thermodynamic calculations of these processes have been made, but no information is provided on what to do with the CO<sub>2</sub> (even the authors note the "harmless" formation of carbon dioxide and water).

The method of separating exhaust gas or smoke formed during fuel oxidation, as well as isolating carbon dioxide from it, is described in patent (Patent No. 2458005, 2012). The flow of exhaust gases is passed through a gas-permeable material, which consists of molecular sieves or activated carbon, then the gas flow containing high-concentration carbon dioxide is separated. The gas with lower carbon dioxide content is released into the atmosphere, while the high-concentration carbon dioxide is used in installations for the production of ammonia, urea, or methanol.

Patent describes a method for extracting CO<sub>2</sub> from flue gases and an installation for this purpose (Patent No. 2513947, 2011). The installation includes a CO<sub>2</sub> adsorbent into which flue gases containing CO<sub>2</sub> are introduced. The CO<sub>2</sub> is contacted with the adsorbent, where it is separated from the flue gases. The installation for extracting CO<sub>2</sub> includes a combustion catalyst (based on metallic Pd or metallic Pt). CO<sub>2</sub> in the enriched solution (absorbed CO<sub>2</sub>) is removed in the regenerator, and the depleted solution is reused separately in the CO<sub>2</sub> absorber.

Work related to the possibility of converting CO<sub>2</sub> into valuable chemical products was conducted as early as the 1970s (Yan Yu, 1976, p. 264; Brednikov, 1975, p. 2588). PdCl<sub>2</sub>-Ni-graphite, Co-Zr, Zn-Cr, Cu-Zn-containing catalysts were used. To obtain the target products, harsh conditions were required: high temperatures and pressures. The authors aimed to soften the conditions for syntheses based on CO<sub>2</sub>. Using a raw material mixture consisting of (mass %): hydrogen – 35, nitrogen – 46, methane – 6, and impurities CO and CO<sub>2</sub> ~13, and industrial catalysts (alumina-cobalt-molybdenum, zinc-chromium, nickel on kieselguhr), they conducted the reaction of CO<sub>2</sub> with H<sub>2</sub> on a flow laboratory unit. The temperature was varied from 200-230°C, the H<sub>2</sub> ratio was (1-3):1 mole, and the pressure was atmospheric. It was found that at a temperature of 200-230°C and a gas flow rate of 120 h<sup>-1</sup>, it is possible to achieve almost complete conversion of carbon oxides to methane (at a molar ratio of hydrogen to carbon oxides of: 1). According to the authors, by excluding the separation and purification unit for the products from the technical scheme, as well as recycling the unreacted hydrogen, the entire resulting mixture can be used as a secondary fuel (methane-hydrogen mixture). The use of an industrial nickel on kieselguhr catalyst allows the conversion of CO<sub>2</sub> to methane under relatively mild conditions: temperature 200-250°C and atmospheric pressure. Non-adsorbed gases from hydrocarbon dehydrogenation processes can be used as a source of hydrogen.

As is known, the basis of a number of alternative methods for producing various hydrocarbons from natural and associated gases, coal, peat, oil shale, and biomass is the Fischer-Tropsch synthesis. Interest in this process remains relevant, as it can solve numerous environmental problems associated with reducing gas emissions into the atmosphere while synthesizing valuable products. The Fischer-Tropsch synthesis of hydrocarbons from carbon monoxide and hydrogen is the second stage of most processes for converting alternative raw materials into synthetic oil and motor fuels. For all processing complexes, it is the most important. In recent years, systematic research has been conducted on Fischer-Tropsch synthesis under the leadership of Academician S. N. Khadzhiev at the Institute of Petrochemical Synthesis of the Russian Academy of Sciences. Various catalytic systems based on transition metals of group VIII have been studied. It was found that in the presence of these catalysts, synthesis gas at atmospheric pressure is mainly converted to methane (Khadzhiev, 2011, pp. 25-32).

Fischer-Tropsch syntheses were carried out on nanoscale particles of an iron-containing catalyst (Patent No. 2485048, 2013). It was shown that the main byproduct in Fischer-Tropsch synthesis (with the use of an iron-containing catalyst) is carbon dioxide, which leads to a significant reduction in the yield of synthetic oil (based on the starting product in alternative raw materials (Khadzhiev, 2011, pp. 84-96). Carbon dioxide can be either a primary or a secondary product in these syntheses. It is noted that in the first case, the formation of CO<sub>2</sub> occurs as a result of the interaction of CO with

surface oxygen formed during the dissociative adsorption of carbon monoxide on the catalyst surface. The carbon from dissociative adsorbed CO participates in the growth of the hydrocarbon chain. The formation of CO<sub>2</sub> as a secondary product occurs during the interaction of CO and water, the primary product of the synthesis. Thus, it is evident from the presented material that by conducting targeted syntheses, the desired result can be achieved and environmental performance improved.

### Conclusion

Climate change and environmental protection issues are among the most urgent issues in modern times. In this regard, the disposal of CO<sub>2</sub> for the purpose of air purification is important. From the point of view of the development of science and technology, it is important to carry out research and studies in these directions.

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