

Adsorption Removal of Benzene from Hydrocarbon Fractions in the Production of Commercial Motor Gasolines

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Abstract

The prospects and features of the implementation of the process of adsorption purification of hydro-carbon fractions from benzene in the production of commercial motor gasoline are considered. It is known that benzene belongs to hazard class I according to the IARC classification, as a result of which its maximum removal from gasoline is one of the main tasks of the oil refining industry. It has been proven that adsorption purification, which is a selective method, is a promising process aimed at ensuring the required quality of automotive gasoline in accordance with current environmental safety standards (Euro 5) due to its simplicity of implementation, high selectivity, and economic and environmental advantages. Benzene removal is a rather complex technical task, but it can be effectively implemented using modern adsorbents such as NaY and ZSM-5 zeolites (selectivity – 80–95 %) and MIL-101 and UiO-66 MOF systems (selectivity – 90–99 %). The main direction of integrating adsorption purification of hydrocarbons from benzene is the possibility of its technological combination with gasoline secondary processing units, catalytic cracking, and reforming within a single automobile gasoline production facility.

Keywords: Automotive gasoline; Hydrocarbon fractions; Benzene; Adsorption purification; Adsorbent; Selectivity; Technology; Regeneration; Octane number.

1. Introduction

The rapid deterioration of the environmental situation in large cities of Ukraine, caused by the expansion of the vehicle fleet, contributes to the introduction of new, stricter environmental safety standards for the chemical composition of motor fuels, in particular automotive gasoline. One of the main hazardous hydrocarbons contained in oil or gas condensate raw materials and capable of entering the composition of their processing products – commercial motor gasoline – is benzene (C₆H₆). The danger of benzene is mainly due to its carcinogenicity (hazard class I) according to the WHO (IARC) classification [1]. When automotive gasoline burns, benzene and phenol vapors are released into the atmosphere and combine with nitrogen oxides to form photochemical smog. Once it enters the lithosphere and hydrosphere, they does not break down easily and can accumulate in ecosystems, poisoning them. At the same time, it is necessary to develop new effective purification systems [2].

Therefore, in order to minimize these negative effects, oil refineries must first control the benzene content in the commercial motor gasoline they produce in accordance with European Union standards [3-4]; and second, develop and implement new effective technologies for removing benzene from hydrocarbon fractions.

2. The objective of the research

Despite its dangerous nature, benzene is a valuable chemical raw material for organic synthesis due to its properties; it is the main representative of aromatic hydrocarbons and serves as a starting compound for the production of many important organic substances.

The benzene molecule can be viewed as a delocalized π -system in which each carbon atom (C) has three σ -bonds (two with neighboring C atoms and one with a hydrogen atom (H)), and the fourth electron of the p-orbital does not participate in σ -bonds, forming π -bonds. Since the C atoms in benzene are located in the same plane, their p orbitals overlap above and below the plane of the ring, forming a single shared electron cloud in which π electrons do not belong to individual bonds but are considered to be distributed throughout the ring [5-7]. The structure of the benzene molecule makes it chemically stable to some reactions but active in electrophilic substitution, allowing various functional groups to be introduced into the molecule. Thanks to this, benzene is a universal platform for the synthesis of compounds such as [8-11]: phenol (C_6H_5OH) – the basis for phenol-formaldehyde resins, nylon, antiseptics; aniline ($C_6H_5NH_2$) – the starting compound for dyes, medicines, polyurethanes; styrene ($C_6H_5CH=CH_2$) – a monomer for polystyrene; toluene ($C_6H_5CH_3$) – a solvent and raw material for explosives (trinitrotoluene); xylenes – components for varnishes, synthetic fibers, PET plastic (via terephthalic acid) and cyclohexane – an intermediate product in the production of caprolactam (nylon-6).

Therefore, given that benzene is a fundamental organic compound underlying the production of most modern synthetic materials, industrial methods for purifying gasoline fractions from benzene must ensure its removal while preserving it for further use. This approach will significantly improve the economic efficiency of the purification process.

3. Results and discussion

Today, in countries such as Ukraine, Japan, Brazil, Canada, India, China, and the European Union, the benzene content in automotive gasoline is limited to ≤ 1.0 % by volume [12-13]. In the US, stricter regulations are in place, limiting benzene content to ≤ 0.62 % [14].

The main sources of total benzene content in commercial automotive gasoline are benzene contained in the straight-run base fraction (0.08–8.0%); benzene contained in FCC gasoline (0.5–1.3 %) and benzene in reforming gasoline – reformate (0.2–8.0 %) [15].

Among the methods for purifying hydrocarbon fractions from benzene, selective purification methods are of particular interest and promise for widespread industrial implementation due to their simplicity (see Fig. 1).

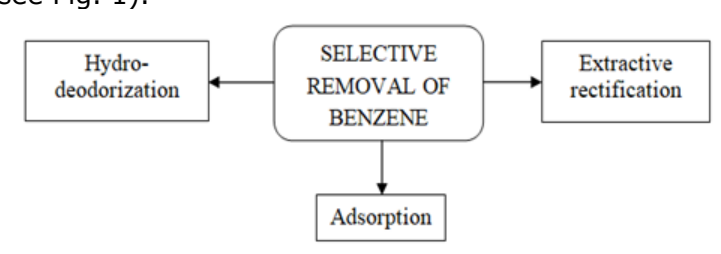


Figure 1. Methods for selective removal of benzene.

Features of methods for selective purification of hydrocarbon fractions from benzene are shown in Table 1.

Table 1. Features of methods for selective purification of hydrocarbon fractions from benzene

Method	The key process	Basic conditions
Adsorption purification	Absorption of benzene by solid adsorbents (zeolites, activated carbon, silica gel) from the gas or liquid phase	Moderate temperature, atmospheric or moderate pressure
Hydrodeodorization	Catalytic hydrogenation of aromatic compounds to alkanes	High pressure (2–10 MPa), high temperature (300–400°C), presence of catalysts
Extractive rectification	Removal of benzene from a mixture using special solvents (extractants) followed by rectification	High temperature, complex equipment scheme

When analyzing methods for the selective purification of hydrocarbon fractions from benzene, it should be noted that adsorption purification has a number of significant advantages over other methods, which are as follows [16]:

- 1) Energy efficiency. Does not require high temperatures and pressures, therefore, lower energy costs.
- 2) No need for hydrogen. Unlike hydrodeodorization, no expensive hydrogen and catalysts are required.
- 3) Simplicity of equipment. Adsorption columns are used, which are simpler and cheaper than high-pressure reactors or rectification units.
- 4) Selectivity. Adsorbents selectively absorb aromatic compounds (especially benzene) without affecting paraffin and naphthenic components.
- 5) Possibility of adsorbent regeneration. Adsorbents can be reused multiple times after benzene desorption (regeneration by heating or gas purging).
- 6) Preservation of raw material composition. Hydrodeodorization changes the chemical composition (converting benzene to cyclohexane), whereas adsorption only removes benzene without altering the other components.
- 7) Environmental friendliness. It does not produce by-products or toxic solvents (as in extractive rectification).
- 8) Renewable. The adsorbent can be regenerated multiple times.

However, this selective purification method has certain disadvantages: limited effectiveness at very high benzene concentrations; the need for periodic regeneration of the adsorbent; unsuitable for complete purification of large flows without cascade systems.

Thus, adsorption purification is a more economical, simpler, and more environmentally friendly method for removing benzene from hydrocarbon feedstock, as it does not require high energy consumption, does not alter the fuel's composition, and does not require expensive reagents or solvents. This method ensures the selective removal of benzene (and partially other aromatic hydrocarbons) from gasoline fractions to a residual content of < 1 %, which meets the requirements of the environmental safety standard Euro-5.

The practical application of this method requires the preparation of hydrocarbon raw materials. Thus, if it is necessary to purify straight-run base fractions (boiling range – 30–180°C), they are first separated into narrower fractions outside the boiling range. Benzene is capable of forming azeotropes with other hydrocarbons, especially low-molecular ones (C₄–C₈) [17–18]. This is primarily due to differences in the interaction between the molecules of the components.

Thus, benzene has strong π – π interactions between aromatic rings, while saturated hydrocarbons have only weak van der Waals forces. As a result, the mixture has a changed saturated vapor pressure, and a common boiling point arises that is lower or higher than that of the pure components: benzene – n-hexane (C₆H₁₄) – boiling point 64.9°C; benzene – n-heptane (C₇H₁₆) – 80.2°C; benzene – cyclohexane (C₆H₁₂) – 79.8°C; benzene – isooctane (C₈H₁₈) – 90.1°C; benzene – toluene (C₇H₈) – 103.3°C.

Benzene azeotropes with hexane, heptane, and toluene complicate the separation of gasoline fractions during rectification [17], therefore, it is impossible to remove benzene from the gasoline fraction solely by rectification aimed at extracting fairly narrow fractions (75–85°C).

Given benzene's ability to form azeotropes, the C₆–C₇ fraction (boiling range ~ 62–110°C) can be considered optimal for its adsorption removal. When cleaning the catalyst and reformate, preliminary preparation may consist of removing water (H₂O content < 1 ppm) to avoid hydrolysis of the adsorbent and filtration/degassing to remove mechanical impurities and gases that can block the pores of the adsorbent [19].

The main industrial adsorbents that have proven themselves well in benzene adsorption processes are activated carbon (e.g. BAC (Benzene Activated Carbon), Norit GAC 1240/830) and zeolites (e.g. 5A, 13X, ZSM-5). At the same time, in recent years, new materials with catalytic or selective properties have emerged as promising alternatives to traditional adsorbents for benzene adsorption purification processes. These materials not only absorb benzene, but can also partially convert it or selectively remove it from other hydrocarbons.

The main groups of alternative adsorbents/catalysts are listed in Table 2.

Table 2. Main groups of alternative adsorbents/catalysts [20-25].

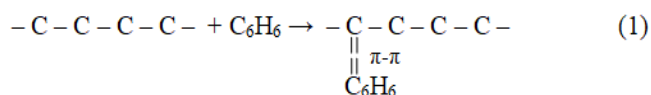
Group of materials	Industrial brands	Characteristics and advantages
Zeolites (molecular sieves)	NaY, HY, β -zeolite	High selectivity for aromatic compounds; possibility of fine tuning pores (5–7 Å); Resistant to high temperatures and regeneration
Metal-organic frameworks (MOFs)	MIL-101(Cr), HKUST-1(Cu) UiO-66(Zr),	Very high surface area (>1000 m ² /g); High selectivity for n -systems (benzene, toluene); possibility of modification with functional groups for selectivity
Mesoporous materials	MCM-41, SBA-15	Controlled pore size (2–10 nm); good adsorption of large molecules; mechanical stability
Metal oxide catalysts	Al ₂ O ₃ , TiO ₂ , ZnO, Fe ₂ O ₃ (sometimes with Pt, Pd)	Capable of not only adsorbing, but also catalyzing partial oxidation or hydrogenation of benzene; used in gas purification systems
Carbon nanomaterials	Graphene, carbon nanotubes (CNTs), carbon aerogels	High surface area and chemical stability; good n - n interaction with aromatic molecules; possibility of modification with metals or oxides to increase selectivity
Functionalized polymer adsorbents	Polystyrene-divinylbenzene, ion exchange resins	Selectivity to aromatic compounds; mild operating conditions, and good regeneration

Therefore, the main advantages of alternative adsorbents can be considered to be higher selectivity to benzene even in the presence of similar hydrocarbons (hexane, heptane), the possibility of recovery (regeneration) without loss of activity; operability in a wider range of temperatures and pressures, characterized by lower energy consumption during desorption, some (MOF, zeolites) allow simultaneous removal and concentration of benzene for further processing. Due to these properties, such adsorbents/catalysts are considered promising materials for environmentally safe and effective purification of gasoline fractions from benzene in industrial conditions.

Considering such high-octane components of commercial gasoline as reformat and catalyst, it should be noted that in terms of the efficiency of benzene removal from their composition, adsorbents can be ranked in the following order: activated carbon (50-90 %) < zeolites NaY and ZSM-5 (80-95 %) < MOF systems MIL-101 and UiO-66 (90-99 %).

The process of benzene removal can proceed either through physical adsorption (purely via intermolecular interactions) or through chemical adsorption (via chemical interactions).

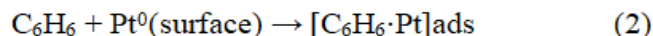
Physical adsorption. The interaction between benzene and an adsorbent (e.g. activated carbon) can be generally represented as follows:



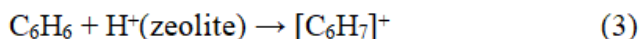
Weak Van der Waals forces arise between the n -electron system of benzene and the surface of the adsorbent. The benzene ring interacts with the porous structure of the adsorbent through n - n interactions. This process is reversible (the interaction energy is quite low, 20–40 kJ/mol) – benzene is capable of desorption when heated or purged with an inert gas. This is the basis for adsorbent regeneration methods.

Chemical adsorption (chemisorption). With this type of adsorption, a chemical interaction occurs between benzene and the functional groups or active centers of the adsorbent. Chemisorption occurs due to the formation of coordination bonds between benzene and metals (Pt, Pd, Cu, Fe) on the surface of catalysts or electrophilic and nucleophilic addition on functionalized adsorbents (in particular, acid-activated zeolites or metal oxides).

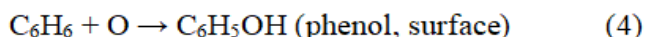
The formation of coordination bonds between benzene and metal on Pd or Pt adsorbents can be represented as follows:



The electrophilic addition of benzene to the acid centers of zeolite should be considered as follows:



The oxidation of benzene on activated metal oxide adsorbents (functionalized MOFs, Pt/C, Pd/C) often occurs at high temperatures and proceeds as follows:



Under industrial conditions, chemical adsorption, like physical adsorption, is often reversible, especially when heated or purged with an inert gas. It should also be noted that physical adsorption of benzene is the most common process in industry, as it is reversible and does not destroy benzene, whereas chemisorption should be used only when benzene needs to be concentrated or converted into other valuable compounds.

The adsorption unit for the adsorption purification of gasoline fractions/components from benzene may consist of the equipment shown in Fig. 2.

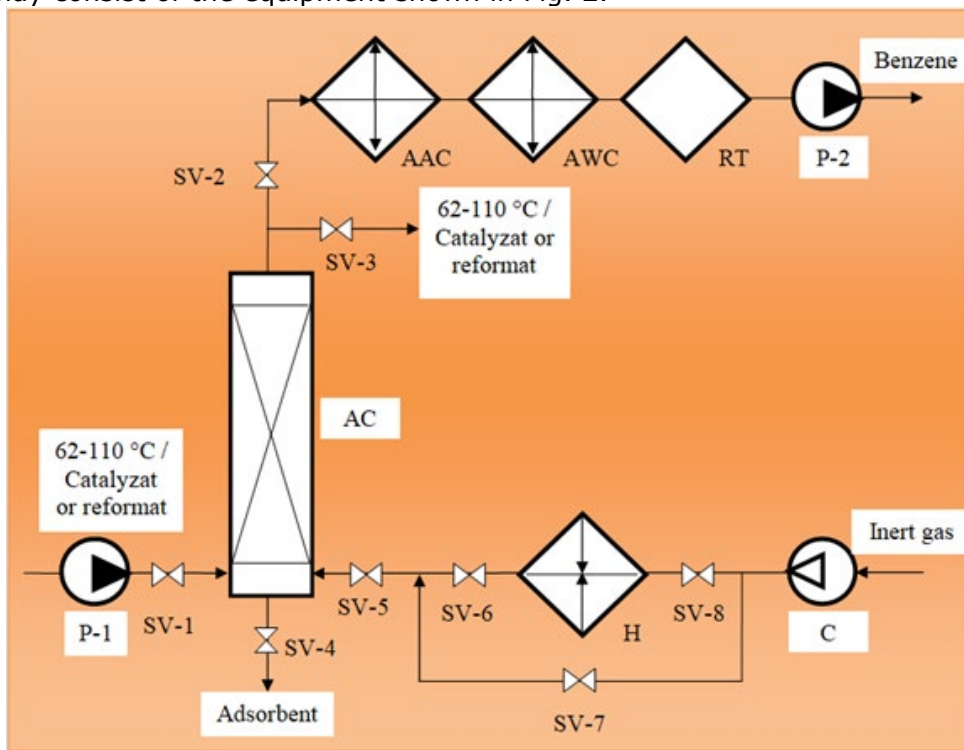


Figure 2. Schematic diagram of the adsorption purification unit for removing benzene from the gasoline fraction.

According to this scheme, the catalyst and reformat from the reforming or catalytic reforming unit, with a temperature of 30–60 °C, are fed by pump P-1 to adsorption column AC, where the pressure is maintained at 1–3 MPa (depending on the type of raw material). The feedstock (62–110°C fraction/catalyst or reformat) passes through a layer of adsorbent. Benzene and other aromatic hydrocarbons are selectively retained in the pores of the adsorbent, and a product with a low benzene content (< 1 %) exits from the top of the AC. Next, after the adsorbent is saturated with benzene and other aromatic hydrocarbons, it is regenerated.

The adsorbent is regenerated directly in the adsorption reactor by passing a stream of inert gas or water vapor (for some catalytic adsorbents) heated to 150–250°C through the adsorbent bed in the heater H. Inert gas is pumped through H using compressor C. To do this, switch the SV-1 shut-off valve, disconnecting the supply of raw materials, and the SV-3 valve, blocking the purified gasoline fraction pipeline. Benzene is removed from the adsorbent, exits from the upper part of the AC, is cooled in the AAC air cooling apparatus, the AWC water cooling apparatus, condenses, and accumulates in the RT reflex tank at a temperature of 25–30°C. Next, benzene is removed from the unit using a P-2 pump for further storage in a tank farm or for processing. To cool the adsorbent, a compressor C is used to blow inert gas at a temperature of 20–25°C through it. To do this, shut off valves SV-6 and SV-8 are closed and valve SV-7 is opened. Continuous operation of the above circuit involves repeated cycles of adsorption → regeneration → cooling.

The main advantages of this scheme include: continuous operation of the unit (due to parallel adsorbers, periodic regeneration is used—one reactor operates while the other regenerates); no mechanical wear of the adsorbent due to constant unloading; automation of the process and minimization of downtime. The adsorbent is discharged using the SV-7 discharge valve from the bottom of the AC in cases where: the adsorbent degrades (e.g. coking); the adsorbent's resource is exhausted (decrease in capacity); for regeneration on a separate stand or disposal. The necessary depth of benzene purification is ensured by analyzing the composition at the inlet and outlet (chromatography), temperature control of the adsorbent layers, manometry (testing the pressure in the column), periodic replacement or renewal of the adsorbent (after dozens of cycles). The integration of the benzene adsorption purification unit into existing commercial gasoline production schemes, which are currently being successfully implemented at oil refineries, is shown in Fig. 3.

According to the diagram shown in Fig. 3, hydrocarbon feedstock – crude oil or gas condensate – is fed into the primary processing unit. In this unit, the raw materials are desalted and dehydrated, and hydrocarbon gases (C₁–C₄) and the broad gasoline fraction (30–180 °C) are removed. Butane (C₄) is separated from the gas mixture by separation or fractionation and sent to the isomerization unit for the production of isomerizate, a high-octane (RON=87–95 points) component of automotive gasoline. The broad gasoline fraction (C₅–C₁₂) with a low benzene content (≤ 0.5%) can be fed directly to the commercial gasoline compounding unit or separated into narrow fractions in the gasoline secondary processing unit.

The C₆–C₁₂ fraction, after the secondary gasoline processing unit, enters the catalytic reforming unit, and after reforming, the finished product – reformat, a high-octane gasoline component (RON = 90–103 points) – enters the adsorption purification unit. In this unit, a catalyst is fed in to extract benzene. This catalyst is a high-octane gasoline component (RON = 85–92 points) formed in the catalytic cracking unit from vacuum gas oil (C₂₀–C₃₀), a product of primary oil refining. Next, the purified reformat and catalyst are fed into the commercial gasoline compounding unit.

If necessary, the narrow fraction (C₆–C₇) from the secondary gasoline processing unit can also be sent to the adsorption purification unit to remove benzene, and then to the compounding unit. This unit also receives high-octane components such as alkylate (RON = 92–99 points) and isomerizate (RON = 85–93 points) and various functional additives (anti-knock, detergent, etc.). After compounding, a stable commercial motor gasoline is obtained that meets current standards for its quality indicators.

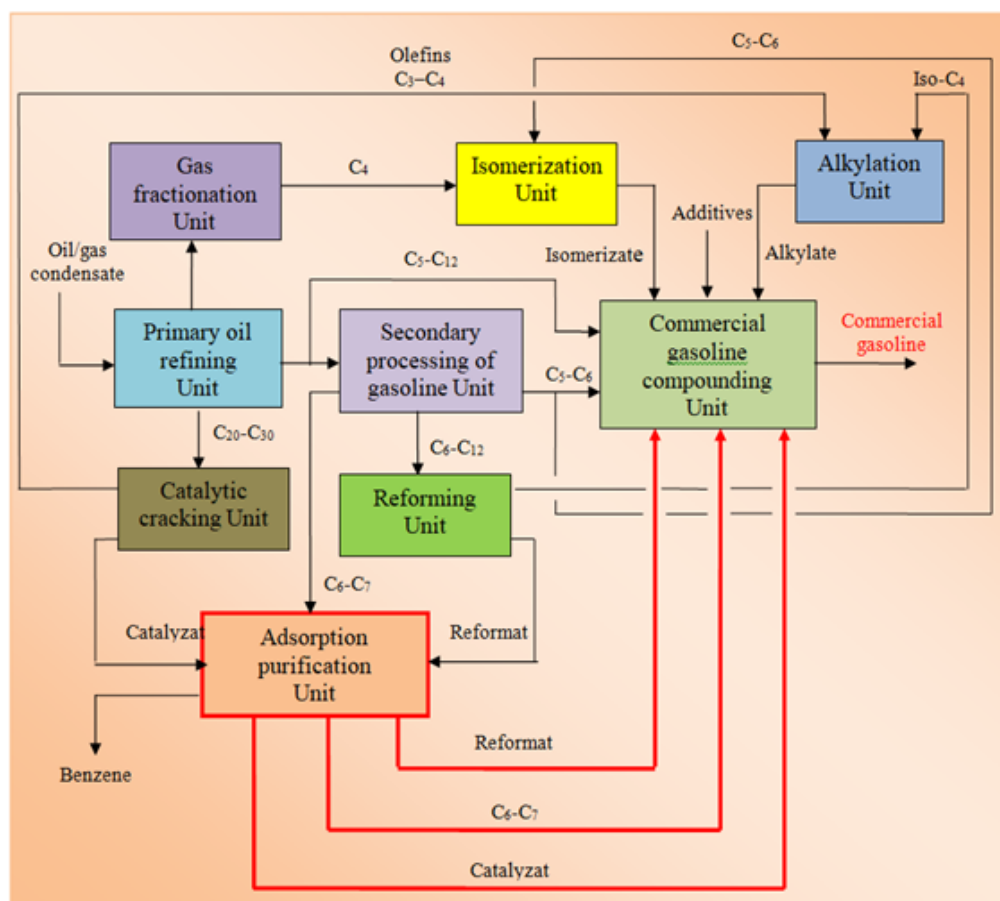


Figure 3. Production scheme for commercial automotive gasoline.

4. Conclusions

Removing benzene from hydrocarbon fractions during the production of commercial motor gasoline is a mandatory step in modern oil refining, as it minimizes the negative impact on human health and the environment during storage, transportation, and use. Today, many countries around the world limit the maximum permissible benzene content in automotive gasoline to 1 %, which is quite difficult to achieve without the use of selective purification methods. The main requirements for these methods are both the depth of benzene removal and its preservation for further processing, since benzene is a valuable raw material for the synthesis of many organic substances – phenol, aniline, styrene, toluene, and xylenes.

Among the methods of selective benzene purification, adsorption purification has quite high prospects for industrial use due to its energy efficiency, simplicity of equipment, high selectivity, and environmental friendliness. Adsorption purification has quite high prospects for industrial use, which can be implemented using modern adsorbents – zeolites NaY and ZSM-5 (selectivity – 80–95 %), as well as MOF systems MIL-101 and UiO-66 (selectivity – 90–99 %).

A positive aspect of using adsorption purification of hydrocarbons from benzene is the possibility of regenerating the spent adsorbent by blowing inert gas or water vapor through it at a temperature of 150–250 °C, directly in the adsorption column without unloading the adsorbent, which ultimately minimizes mechanical wear of the adsorbent due to constant unloading, as well as ensures automation of the process and minimization of downtime. In other words, the operation of benzene adsorption purification plants involves repeated cycles of adsorption → regeneration → cooling.

The main direction of integrating adsorption purification of hydrocarbons from benzene is the possibility of its technological combination with gasoline secondary processing units, catalytic cracking, and reforming within a single facility for the production of automotive gasoline. Compliance with current environmental safety standards for benzene content in commercial motor gasoline produced at oil refineries in Ukraine contributes to increasing the competitiveness of products on the international petroleum products market.

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