

# Synthesis of methylpyridines by catalytic method in the gas phase

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**Abstract.** The article presents the results of the yield of pyridine derivatives obtained on the basis of a catalyst of various compositions and, at the same time, the efficiency of new catalytic systems for the synthesis of pyridine derivatives is investigated. “Angren kaolin” (Uzbekistan) was used to develop catalytic systems. The development and implementation of new catalytic systems leads to a decrease in the problems associated with the purchase of catalysts from abroad and will entail economic benefits for the republic. To solve the problems, the following research methods were used: spectral analysis, microscopic analysis, potentiometric method, physicochemical methods of analysis, and methods of preparing catalysts. The catalysts have been developed with different ratios of starting components. The efficiency of heterogeneous catalysts was determined by the synthesis of pyridine derivatives at the facilities of JSC “Navoiyazot”. It was found that among the monocomponent catalysts, the CK-13 brand catalyst ((CdO-13.0%, kaolin-87.0%) (2-methylpyridine-41.2%, 4-methylpyridine-22.4%) was more efficient) with a yield of 63.6%, and among two-component catalysts of the CChK-13 brand (CdO-13.0%, Cr<sub>2</sub>O<sub>3</sub>-5.0%, kaolin-82.0%) (2-methylpyridine-45.4%, 4-methylpyridine - 24.8%) with a yield of 70.2%. And also the influence of peptizers on the yield of finished products was studied.

## 1 Introduction

Pyridine and its derivatives are widely used in the production technology of surface-active substances (surfactants), rubber vulcanization accelerators, ion-exchange resins, drugs in pharmaceuticals, pesticides, agriculture, starting compounds in the synthesis of films and photographic materials, corrosion inhibitors, high-quality synthetic rubbers, dyes for the textile industry, intermediate products of the synthesis of vinylpyridines and extractants [1-3].

The synthesis of pyridine and its derivatives based on acetylene and ammonia in the presence of a heterogeneous catalyst is considered one of the most promising methods. In the synthesis of pyridine derivatives, catalysts from salts and oxides of copper, mercury, zinc, cadmium, chromium, and iron prepared by various methods are widely used [4-5].

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The advantage of this method is the simplicity of technological conditions and the possibility of starting production on the basis of JSC “Navoiyazot”. Since there is a high demand for pyridine derivatives in the world, including in the Republic of Uzbekistan.

It is known that the process of synthesis of pyridine and its derivatives from acetylene is influenced by many factors, such as the nature of the catalyst, peptizers, temperature, the ratio of starting materials, sorption of reagents and absorption, desorption, dehydrocyclization, and pressure [6–9].

The creation of selective catalysts in the synthesis of pyridine derivatives is one of the urgent problems of chemistry and chemical technology.

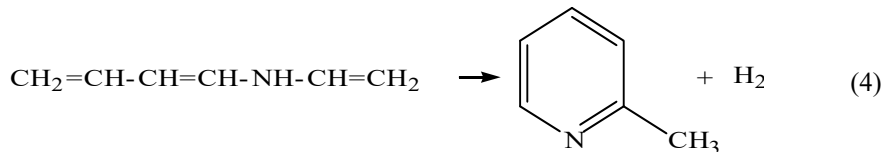
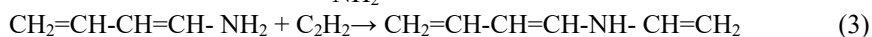
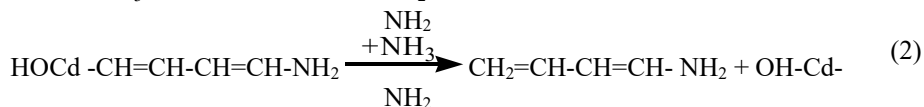
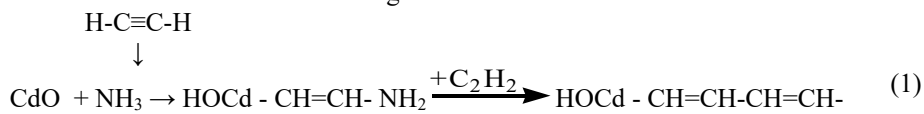
In this research work, catalysts were developed containing a monoactive component: CdO-kaolin, Cr<sub>2</sub>O<sub>3</sub>-kaolin, ZnO-kaolin, and an inactive component: CdO-Cr<sub>2</sub>O<sub>3</sub>-kaolin, CdO-ZnO-kaolin, Cr<sub>2</sub>O<sub>3</sub>-ZnO-kaolin, by peptization of the kaolin carrier with various minerals and organic acids. The brand AKF-78 of kaolin mineral was chosen as the carrier. The properties of mono- and diactive catalysts containing cadmium, chromium, and zinc oxides have been studied. The process of sorption, desorption and dehydrocyclization of acetylene and ammonia on the catalyst surface has been studied.

The influence of the nature and composition of catalysts, peptizers, temperature and volume ratio of reagents on the synthesis of methylpyridines obtained in the process of acetylene amination on the basis of prepared catalysts has been determined. The prepared catalysts are selective catalysts in the synthesis of pyridine derivatives.

The composition of the catalyzate obtained by the reaction of acetylene with ammonia at high temperature includes a mixture of compounds: 2- and 4-methylpyridines, acetonitrile, vinylpyridine, higher representatives of pyridine derivatives - such as lutidines, collidines, various resins and other nitrogen-containing substances [10–11].

In the reaction of acetylene with ammonia, vinylamine is first formed, and then the heterocyclization stages take place:

The reaction is based on the following mechanism:

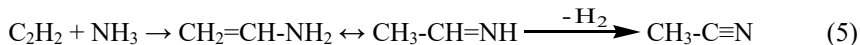


Nucleophilic substances – the ammonia pass into the acetylene at high temperature through compounds of cadmium (II), chromium (III) with aprotic acid centers. The electron-withdrawing properties of these compounds decrease at high temperatures. The nucleophile formed as a result of the separation of the ammonia proton in the reaction centers of the catalysts binds to the metal, and the metal oxides pass into the state CdO+NH<sub>3</sub>→HO-Cd-NH<sub>2</sub>. The ammonia is well sorbed in metal-containing centers [11-12].

At the stage of dehydrocyclization, heterocyclic compounds are formed.

In the cadmium-chromium catalyst, chromium oxide acts as a promoter of dehydrogenation and heterocyclization processes in the synthesis of pyridine derivatives from intermediate vinylamine.

The zinc compounds increase the conversion of vinylamine to ethylamine and then to acetonitrile:



It has been established that on cadmium, chromium, and cadmium-chromium catalysts, the process shifts towards the selective production of 2- and 4-MP. An increase in the formation of acetonitrile was observed in catalysts based on zinc oxide. The using of d-metal oxides, mainly cadmium oxide, increases the activity of reaction catalysts towards the formation of pyridine bases, i.e., heterocyclization reactions. The catalytic activity of cadmium compounds and the ability to adsorb acetylene and ammonia depend on the formation of  $\pi$ -bonds and  $\sigma$ -ligands at high temperature. The presence of the d layer in the  $\text{Cd}^{2+}$  ion plays an important role in the activation of acetylene.

Because of the high melting point, cadmium compounds act as a selective catalyst in the process. As a result of the dehydrocyclization of the raw materials during the synthesis, the yield of the vinylamine intermediate product increases. As a result, methylpyridines are synthesized [12-13].

## 2 Materials and methods

In industry, the main starting materials for the production of pyridine derivatives are acetylene, aldehydes, ketones, hydrocyanic acid, ammonia, and catalysts of various compositions, which are the objects of this work.

The paper considers the influence of cadmium oxide CdO and the mineral kaolin with different active components: ChC-13 ( $\text{Cr}_2\text{O}_3$ -13.0% + kaolin-87.0%), ZK-13 (ZnO-13.0% + kaolin-87.0 %), CChK-13 (CdO-13.0% +  $\text{Cr}_2\text{O}_3$ -5.0% + kaolin-82.0%), CZK-13 (CdO-13.0% + ZnO-5.0% + kaolin-82.0%), ChZK-13 ( $\text{Cr}_2\text{O}_3$ -13.0% + ZnO-5.0% + kaolin-82.0%).

The following research methods were used to solve the set problems: physicochemical methods of analysis, spectral analysis on a Bruker Invenio S-2021 Fourier spectrometer in the range 4000 – 400  $\text{cm}^{-1}$ ; microscopic analysis on a scanning electron microscope of the SEM EVO MA 10 analytical complex and a potentiometric method. The mass fraction of pyridine derivatives was measured by the chromatographic method [14-17].

The main controlled method for the quantitative formation of pyridine derivatives in practice is the potentiometry method. The potentiometric method measures the potential difference (EMF) of two electrodes - measuring and auxiliary, placed in the solution to be investigated. All devices are equipped with a temperature compensator, controlled manually or automatically, since the EMF value is directly dependent on temperature.

According to the method of potentiometric titration, the mass fraction of pyridine and pyridine derivatives in percent is calculated using formula 6.

$$X = \frac{V \cdot 0,07910 \cdot 100}{m} \quad (6)$$

The method of mixing dry components was used to prepare the catalysts.

A certain amount of cadmium oxide (TU 6-09-02-480-89) and chromium oxide (TU 6-09-4272-84) was added to 150 g of kaolin. In order to increase the mechanical strength, as well as to create acid sites on the catalyst surface and to ensure a uniform distribution of components in kaolin, 50 ml of phosphoric acid solution and 85% of distilled water were added to the resulting mass until a pasty mass was obtained.

The resulting homogeneous mass was formed through an extruder with a diameter of 2 mm, then the resulting cylinders of catalyst samples 4 mm long were sieved from dust and dried at a temperature of  $100 \pm 5$  °C for 3 hours, then calcined at a temperature of  $600 \pm 25$  °C

with a temperature rise of 50 °C per hour while maintaining a temperature of about 600 °C for 3 hours. The resulting finished catalyst has a composition, wt %: CdO-13.0%, Cr2O3-5.0%, kaolin-82.0% [16].

Determination of the Mass Fraction of Catalyst Losses on Ignition

Appliances, utensils, reagents:

Laboratory scales for general purposes - according to GOST 24104 of the second accuracy class with the maximum weighing limit of 200 g.

Weights G-2-210 - according to GOST 7328.

Laboratory resistance electric furnace SNOL-1,6,2,5.1/11-IZ - according to TU 16-681.139 or any other, providing a heating temperature of (900 ± 10) °C.

Desiccator 1-140 - according to GOST 25336, filled with calcium chloride, previously calcined at a temperature of 250-300 °C for two hours.

Crucible low 3 - according to GOST 9147.

Calcium chloride (calcium chloride) dehydrated - according to TU 6-09-4711.

1.0000 g of finely ground catalyst is weighed in a crucible calcined to constant weight and weighed with the same accuracy, then the catalyst is placed in an electric furnace and calcined at a temperature of (750 ± 10) °C, kept at this temperature for 2 hours, cooled in desiccator and weighed with the same accuracy. The calcination is repeated until constant weight.

The mass fraction of losses of the catalyst during ignition ( $X_3$ ) as a percentage is calculated using formula 7:

$$X_3 = \frac{m - m_1}{m} \cdot 100 \quad (7)$$

There  $m$  is the weight of the catalyst sample before calcination, g;

$m_1$  is the weight of the catalyst sample after calcination, g.

The result of the analysis is taken as the arithmetic mean of the results of two parallel determinations, the absolute discrepancy between which should not exceed 0.2%.

$m$  is the weight of the catalyst sample, g.

$X_3$  is the mass fraction of catalyst losses during calcination, %.

The average arithmetic mean of the results of two parallel determinations is taken as the result of the analysis, the absolute discrepancy between which should not exceed 0.25%.

Analysis methodology

The mass fraction of pyridine derivatives was measured by the chromatographic method.

The duration of the analysis is 30 - 40 minutes.

The principle of the chromatographic method is based on the separation of the mixture under study into its constituent components in a chromatographic column, the conversion of measurements of physical and chemical properties in an ionization-flame detector into an electrical signal, regmetering and measurement of the results of this separation.

Auxiliary devices, reagents and materials:

Laboratory gas chromatograph of any type with an ionization-flame detector. Separating metal column, 3 m long, with an inner diameter of 3 mm.

Mechanical stopwatch SOS, pr-26-2-000, GOST 5072-79E;

Caliper, GOST 166-89;

Microsyringe, type MSH-10, TU 2-833.106;

Foam gas flow meter;

Vacuum pump of any type;

General purpose laboratory scales - accuracy class model VLR - 200, GOST 24104 - 88E or another type with the smallest weighing limit of 200 g and a pressure price of  $0,5 \cdot 10^{-3}$  g.

Set of weights G-2-210, GOST 7328-82E.

Penicillin bottle with self-sealing rubber stopper;

Gaseous nitrogen in a cylinder, GOST 9293-74.

Hydrogen gaseous in a cylinder, GOST 3022-80.

Grade A or from a hydrogen generator of any type;  
Air for supplying pneumatic devices, automation equipment, GOST 17433 - 80;  
Sorbent: polychrome (impregnated with liquid phase 1,2,3 - trissbeta - cyanethoxy) propane, 15% by weight of polychrome.

Rectified technical ethyl alcohol GOST 18300-87;

Derivatives of pyridine;

Distilled water, GOST 6709-72.

Before filling, the column is thoroughly washed with ethyl alcohol and blown with air; the column is filled with a sorbent using a vacuum pump.

The filled column is installed in the thermostat of the chromatograph, and not connected to the detector, it is condensed in a stream of nitrogen carrier gas for 6-8 hours at a temperature of 120°C. At the end of conditioning, the column is connected to the detector and the gas lines are checked for leaks by washing.

Thermostat temperature 70 - 80°C

Evaporator temperature 150°C

Volume flow rate of carrier gas (nitrogen) 29 m<sup>3</sup>/h

Volumetric flow rate of hydrogen and air, respectively, 2420 dm<sup>3</sup>/h

The volume of the input analyzed sample and calibration mixture is 1 l

Chart tape speed 240 mm/h

Preparation of calibration mixtures:

Graduation mixtures are prepared in penicillin flasks with self-sealing stoppers.

Mixtures for acetonitrile, pyridine derivatives, ketones, vinylamine in a mixture for acetone - in water.

To do this, 5 sm<sup>3</sup> of a solvent (acetone or distilled water) is introduced into a pre-weighed penicillin flask, weighed, then the calculated amount of the required component is introduced into the flask, and weighed again.

The prepared calibration mixture is thoroughly mixed by shaking.

The sample to be analyzed is injected three times into the evaporator of the chromatograph using a microsyringe. The heights of the peak analyzed component on the chromatogram are measured and their average value is calculated [17-18].

Technological scheme for the synthesis of picolines (pyridine derivatives).

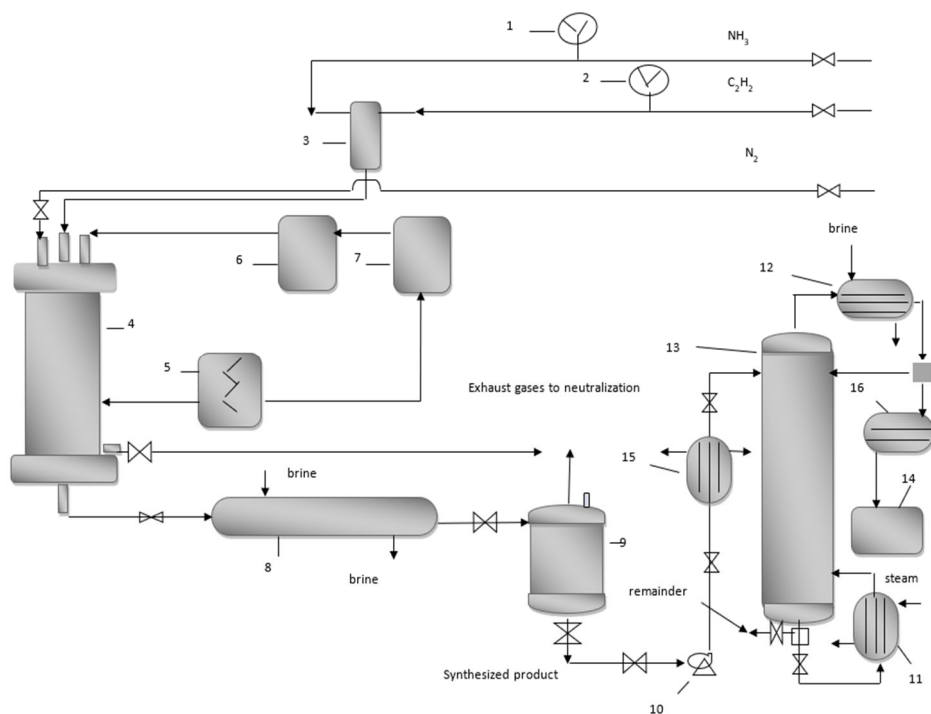
A technological scheme for the synthesis of picolines has also been developed and is presented in Figure 1.

In the reactor 4 is placed 100 g of the prepared sample of the catalyst. After that, the reactor is connected to the electrical source 5 and the reactor starts to heat up. When a temperature of the reactor reaches 300 °C, acetylene and ammonia are blown from the gas pipeline on both sides of the mixer 3, while the gas flow rates are measured by the rheometer 1,2 and the gases are mixed in the mixer 3, and the resulting mixture of gases from the bottom of the mixer 3 is fed into the reactor 4 with a hose. The experiment is carried out in the temperature range of 300-420°C.

The synthesis process takes place in the reactor, and the synthesized product in the form of a gaseous substance from the lower part of the reactor 4 enters the heat exchanger 8.

In the heat exchanger, the gases are cooled by brine and begin to accumulate in the intermediate tank 9.

The synthesized substance is fed by the intermediate capacity 9 with the help of a centrifugal pump 10 to the heat exchanger 15, where it is heated to the boiling point. The heated mixture is fed to the separation in the rectification column 13.



1,2-rheometer; 3-mixer; 4- reactor; 5-electric heater; 6-thermocouple; 7-potentiometer; 8,15,16 - heat exchangers; 9- intermediate capacity; 10-pump; 11-boiler; 12-reflux condenser; 13- rectification column; 14 - capacity of finished products.

**Fig. 1.** Principal technological scheme for the synthesis of picolines.

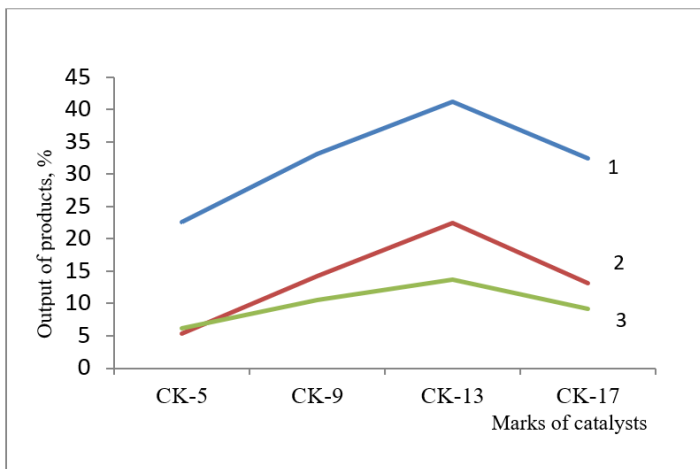
Flowing down the column 13, the liquid interacts with the rising vapor, which is formed when the bottom liquid boils in the boiler 11. As a result of mass exchange with the liquid, the vapor is enriched with a volatile component. For a more complete enrichment, the upper part of the column 13 is refluxed, which is obtained in the dephlegmator 12 by condensing the steam leaving the column 13. Part of the condensate is removed from the dephlegmator 12 in the form of a finished product of separation - distillate, which is cooled in the heat exchanger 16 and sent to the intermediate capable 14.

### 3 Results

During the condensation of acetylene and ammonia, the following substances are mainly synthesized: 2- and 4-methylpyridine (2- and 4-MP), 2-methyl-ethylpyridine, acetonitrile, 2,4,6-trimethylpyridine.

To study the yield of finished products, we have developed and studied catalysts based on local components containing a monoactive component: CdO-kaolin with various ratios at the temperature of 420 °C (Figure 2).

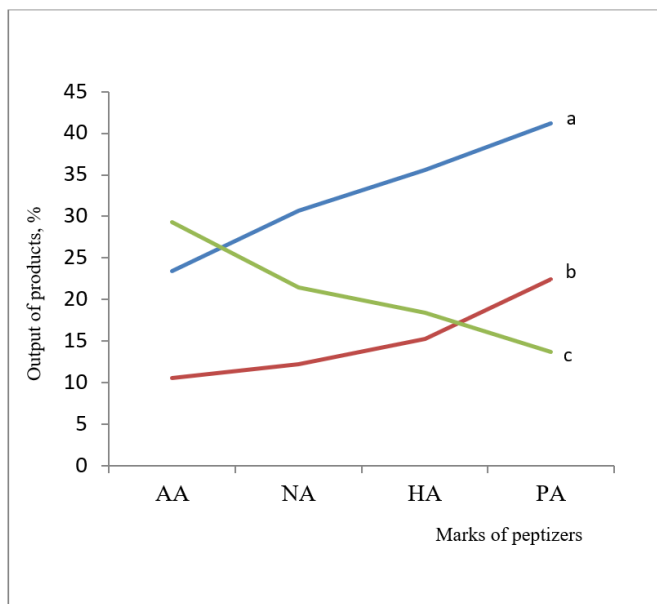
The results of the dependence of the yield of finished products on the nature and composition of the monoactive catalyst are presented in the Figure 2.



**Fig. 2.** Dependence of the yield of methylpyridines on the nature of the catalyst (temperature 420 °C): 1-2-MP; 2-4-MP; 3-other connections.

Results in Figure 2 show that the formation of methylpyridines depends on the amount of the active component of cadmium oxide CdO in the catalyst. Amounts of the active CdO component were tested ranging from 3.0% (MP-18.5%) to 13.0% (MP-63.6%). An analysis of the experiments shows that the most active catalyst is cadmium oxide CdO with a percentage of 13.0% and, at the same time, methylpyridines 63.6%, respectively, 2-MP(2-methylpyridine)-41.2%,4-MP(4- methylpyridine) – 22.4%.

Peptizers used in the preparation of heterogeneous catalysts are of great importance for the yield of the finished product.



**Fig. 3.** Dependence of the yield of methylpyridines on the nature of the peptizers (AA- acetic acid; NA-nitric acid; HA- hydrochloric acid; PA- phosphoric acid) (temperature 420°C): a-2-MP; b-4-MP; c-other connections.

When catalysts are treated with mineral and organic acids, their acidity centers increase. The increase in acid centers enhances the process of heterocyclization of acetylene. Therefore, the yield of finished products also depends on the use of peptizers. Peptizers enhance the interaction of active components and nuclei. As a result, release of peptizers in the state of various concentrations during the firing of the prepared catalysts increases the pores in the catalysts, and this, in turn, ensures the increase in the active surface of the catalyst.

Figure 3 shows the results of the preparation of catalysts during the peptization of various mineral and organic acids at the temperature of 420°C.

The results of the experiment show (table 2) that when preparing monoactive catalysts with organic acid CH<sub>3</sub>COOH (with concentrations of 90%), the yield of finished products (MP -34.0%) is relatively low, and when using mineral acids HNO<sub>3</sub> (with a concentration of 56.0%), HCl (with a concentration of 20.0%) and H<sub>3</sub>PO<sub>4</sub> (with a concentration of 85.0%) as peptizers and the preparation of catalysts based on them provides the formation of methylpyridines in high yield (MP-42.9 %, 50.9%, 63.6% respectively).

Also, different brands of catalysts with mono- and inactive compositions were prepared and catalytic properties were studied in the temperature range from 340÷460 °C (table 1).

For example, when carrying out the synthesis in the temperature range of 340–420°C using a catalyst of the brand CCK-13 (CdO-13.0% + Cr<sub>2</sub>O<sub>3</sub>-5.0% + kaolin-82.0%), the yield of 2-methylpyridine increases from 35.7 to 45.4%, 4-methylpyridine from 15.4 to 24.8% (in the amount of 2- and 4-methylpyridines 70.2%), respectively, with the increasing temperature.

With the increasing temperature from 440 °C, the increasing in the yield of nitrogen-containing compounds (vinylpyridine, higher representatives of pyridine derivatives, lutidines, nitriles, various resins) is observed. And this leads to poisoning of cadmium oxide, which is considered the main active ingredient.

**Table 1.** The dependence of the yield of methylpyridines on the temperature.

№	Catalysts' name	Temperature, °C	Synthesized products, %		
			2-MP	4-MP	Other connections
1.	CK-13. (CdO-13.0%, kaolin-87.0%)	340	28.6	9.4	5.1
		360	30.7	12.3	6.4
		380	35.3	16.7	8.6
		400	38.6	19.5	10.2
		420	41.2	22.4	13.7
		440	36.3	17.6	18.4
		460	30.4	11.3	23.6
2.	ChK-13. (Cr <sub>2</sub> O <sub>3</sub> -13.0%, kaolin -87.0%)	340	24.6	7.3	6.4
		360	26.7	8.8	9.1
		380	27.4	12.3	10.5
		400	29.4	14.6	13.8
		420	32.5	16.7	15.8
		440	27.5	14.4	19.9
		460	23.3	8.8	25.5
3.	ZK-13. (ZnO-13.0%, kaolin-87.0%)	340	19.4	6.6	10.1
		360	22.3	7.5	13.4
		380	23.3	9.4	15.8
		400	25.7	11.5	19.8
		420	27.5	13.3	22.3
		440	25.4	11.5	28.1
		460	20.1	4.4	33.6
4.	CKhK-13.	340	35.7	15.4	3.6
		360	37.2	17.4	5.3



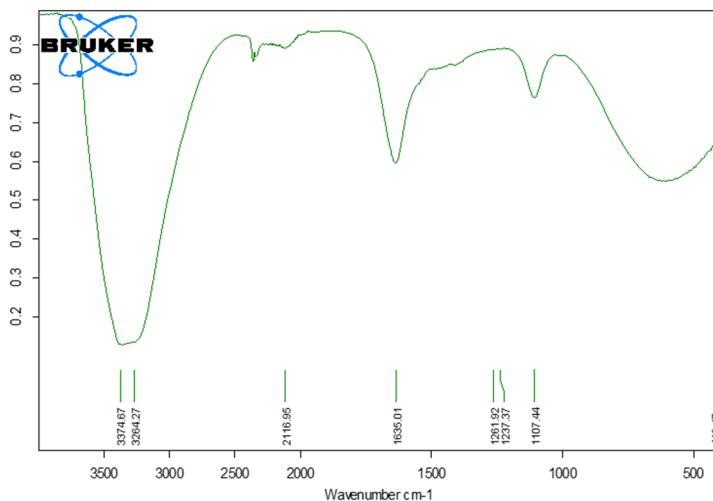
	(CdO-13.0%, Cr <sub>2</sub> O <sub>3</sub> -5.0%, kaolin-82.0%)	380	39.5	19.8	6.4
		400	43.2	22.6	8.8
		420	45.4	24.8	10.2
		440	43.5	22.2	12.3
		460	41.8	20.8	14.7
5.	CZK-13. (CdO-13.0%, ZnO -5.0%, kaolin-82.0%)	340	29.6	8.4	5.2
		360	32.0	11.5	7.6
		380	35.5	13.0	9.7
		400	38.4	16.6	11.5
		420	41.8	19.2	13.2
		440	39.2	16.2	16.5
6.	ChZK-13. (Cr <sub>2</sub> O <sub>3</sub> -13.0%, ZnO-5.0%, kaolin-87.0%)	460	36.5	14.5	19.7
		340	21.4	3.1	3.4
		360	25.0	5.3	6.0
		380	30.7	7.6	9.8
		400	34.9	9.6	13.3
		420	38.7	12.3	16.4
		440	34.3	10.5	19.3
		460	30.8	7.6	21.6

And in order to reactivate the catalyst center, it is treated with phosphoric acid and at the same time the acidity center of the catalyst increases and the catalytic property is restored.

As a result of dehydrocyclization in the reaction of acetylene with ammonia, the yield of pyridine derivatives increases. At a high temperature of 360-420°C, the conversion and adsorption of acetylene increase. It was also shown that the alternative temperature for the formation of methylpyridines (2-MP and 4-MP) using mono- and diactive catalysts is 420°C. Above this temperature, the transition of the active CdO component to the state of metallic cadmium is accelerated, and this leads to the decrease the catalytic activity of the catalyst.

Tab.1 shows that the yield of finished products is higher for catalysts with inactive components compared to monoactive ones.

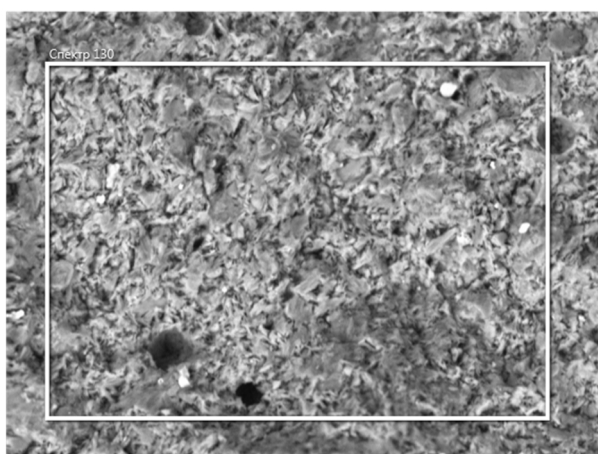
Chromium oxide in the composition of the cadmium-chromium catalyst acts as a promoter of dehydrogenation and heterocyclization processes in the synthesis of pyridine derivatives from intermediate vinylamine.



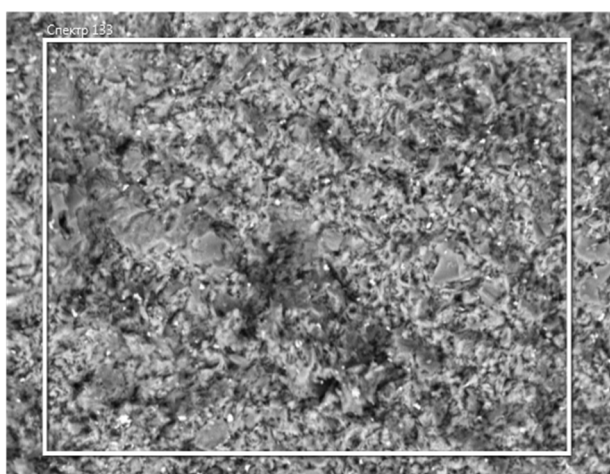
**Fig. 4.** IR-spectrum of the synthesized sample 2- and 4-MP.

Also, to confirm the structure of the synthesized products of pyridine derivatives, the IR spectrum of 2- and 4-MP was obtained (Figure 4).

In the IR spectrum of the synthesized sample of 2- and 4-MP based on the CK-13 catalyst, there is a weak line of the stretching vibration of the primary amine (C-NH<sub>2</sub>) at 3384.47 cm<sup>-1</sup> v, the stretching vibration of the group (N-H) in ethylimine at 3265.48 cm<sup>-1</sup> v and the stretching vibration of the group (C≡N) of acetonitrile at 1992.61 cm<sup>-1</sup> v. Also in the spectrum there is a vibration of the group (N=C) at 1634.53 cm<sup>-1</sup> v belonging to the ring of pyridine derivatives and a plane-strain vibration of the group (C-H) in pyridine derivatives at 1102.26 cm<sup>-1</sup> δ (Figure 2). Based on the IR spectrum, it was proved that the synthesized product is a sample of 2- and 4-MP.



a)



b)

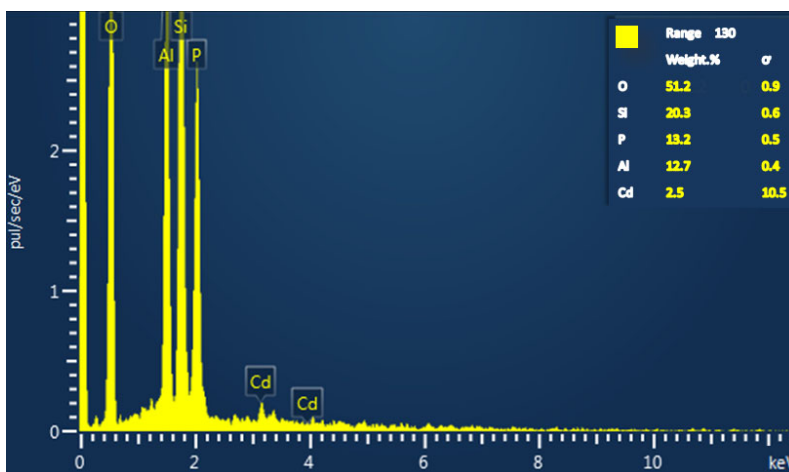
**Fig. 5.** Microscopic images of the prepared catalyst of the brand CK-5 in the size of 50 μm (mkm): a) before application; b) after application.

In addition, images were obtained on a scanning electron microscope with the smallest increase in the surface of the prepared catalyst of the brand CK-5 before and after using in the synthesis process of pyridine derivatives, the composition of cadmium oxide and kaolin, respectively, with percentages of 5 and 95%, and the elemental composition of the catalyst of the brand CK-5 was determined which is before and after application in the synthesis process, the results of which are presented in Figure 5.

Figure 5-a shows the presence of a pore on the surface of the prepared catalyst before use in the synthesis process in the scanned area. In these pores of the catalyst, the adsorption process takes place and after that the synthesis process.

Figures 5-b shows the absorption of the adsorbate on the surface of the prepared catalyst.

And all this proves the semi-quantitative elemental composition obtained from the surface of the prepared catalyst of the brand CK-5 (Figure 6).



**Fig. 6.** Image from the surface of the semi-quantitative elemental composition of the prepared catalyst of the brand CK-5.

## 4 Conclusion

In this work, catalysts with different compositions for the yield of pyridine derivatives were developed and studied. It has been established that among the monocomponent catalysts, the most effective is the CK-13 catalyst ((CdO-13.0%, kaolin-87.0%) (2-methylpyridine-41.2%, 4-methylpyridine-22.4%)) with a yield of 63.6%, and among two-component catalysts - brand CChK-13 (CdO-13.0%, Cr<sub>2</sub>O<sub>3</sub>-5.0, kaolin-82.0%) (2-methylpyridine-45.4%, 4-methylpyridine-24.8%) with a yield of 70.2%.

The peptizers were used in the preparation of heterogeneous catalysts based on various mineral and organic acids. It was found that the use of phosphoric acid as peptizers contributed to the increase in the activity of the catalyst.

Also, cadmium, chromium, and zinc compounds were used as an active component in the composition of the catalyst in the synthesis of methylpyridines. It was found that the yield of methylpyridines increased when using cadmium compounds.

Also, the structures of the synthesized products of pyridine derivatives were proved by the IR spectroscopic method.

The surface images and semi-quantitative elemental composition of the prepared catalyst of the brand CK-5 were obtained and studied.

The technological scheme for the synthesis of pyridine derivatives was developed.

## References

1. G.D. Henry, *Tetrahedron Lett.* **60(29)**, 6043-6061 (2004)
2. E.F.V. Scriven, J.E. Toomey, R. Murugan, *In Kirk-Othmer Encyclopedia of Chemical Technology, Pyridine and pyridine derivatives*. In: Kroschwitz JI, Howe-Grant M (eds), 20. (Wiley, New York, 1996)
3. Y.S. Higasio, T. Shoji, *Appl Catal* **221**, 197-207 (2001)
4. M.K. Nazarbekov, *Synthesis and technology for obtaining pyridine, 2- and 4-methylpyridines from acetylene, tetrahydrofurfuryl alcohol and ammonia* (Tashkent, 1999)
5. A. Ikramov, S.D. Kholikova, SH.A. Ikramova, *The synthesis of pyridine bases based on acetylenic alcohols. Chemistry and chemical engineering* (Tashkent, 2018)
6. A. Ikramov, H.I. Kadirov, S.Zh. Khalikova, N.H. Musulmonov, Sh.A. Ikramova, *DAN ANRUz* **1**, 49-53 (2016)
7. A. Ikramov, M.G. Ismatullayeva, K.M. Akhmerov, *Synthesis of pyridine bases by lamination of acetylene in the presence of ketones* (Tashkent, 1979)
8. S.E. Nurmanov, A. Ikramov, T.S. Sirlibayev, *Heterogeneous-catalytic synthesis of pyridine bases from acetylene, ketones and ammonia in the book: Synthesis, properties and application of organic compounds* pp. 49-52 (Tashkent, 1990)
9. A. Ikramov, S.J. Khalikova, N.KH. Moslemanov, H.I. Kadirov, D.A. Khandamov, *Kimi va technologies kimi* **1**, 23-26 (2017)
10. A. Ikramov, *Synthesis of pyridine bases by catalytic reactions of acetylenic and carbonyl compounds, development of their technology* (Tashkent, 1998)
11. Kh. Vapoev, A. Umrzakov, S. Kodirov, *Synthesis of picolines based on monocomponent catalysts*, International Conference "Integrated Innovative Development of the Zaravshan region: achievements, problems and prospects", Novai 318-319 (2022)
12. Kh. Vapoyev, A. Umrzokov, S. Kodirov, *The impact of the nature of catalysts and peptizers on the synthesis of methyl pyridines* (Universum, technical sciences, 2022)
13. S.Sh. Sharipov, K.S. Sanakulov, B.F. Mukhiddinov, A.T. Umirzakov, *Mining notice of Uzbekistan* **4(83)**, 105-109 (2020)
14. B.N. Tarasevich, *IR spectrum of the main classes of organic compounds* (Moscow, 2012)
15. E.N. Shapovalova, A.V. Pirogov, *Chromatographic methods of analysis* (Moscow, 2017)
16. N.S. Gerasimova, *Potentiometric methods of analysis: Guidelines for homework in analytical chemistry* (Moscow, 2010)
17. Yu.Yu. *Lurie Handbook of analytical chemistry* (Moscow, Chemistry, 1989)
18. GOST 13647-78 Reagents. Pyridine. Technical specifications (with Change N 1) GOST No. 13647-78 dated April 20 (1978)