

# Increasing the production of gas condensate by using ammonium carbonate salts

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**Abstract.** The work is devoted to the problem of increasing gas condensate production in gas condensate fields. It was found that ammonium carbonate salts, in the absence of calcium chloride type waters, interact with carbonate rocks, increase the permeability of reservoirs. Solutions of ammonium carbonate salts when interacting with formation water of the calcium chloride type form chemically precipitated chalk in the pore space, while the permeability of carbonate rocks decreases. A set of experimental studies was carried out to study the displacing and washing properties of ammonium carbonate salts. It was found that ammonium carbonate salts have high displacing properties, the displacement ratio of kerosene by  $NH_4HCO_3$  solution is 0.75-0.80, while reservoir water – 0.55-0.58. According to the results of laboratory studies of the displacing and washing characteristics of ammonium carbonate salts, conclusions were made about the effect of bicarbonate solution (ammonium carbonate salts) on the production characteristics of a well in reservoir conditions at temperatures of 80-100 °C and above. Industrial tests of ammonium carbonate salts showed an increase in gas flow by 30-50% at wells № 23 of Opishnia, № 115 of Mashivka, № 3 of Tymofiivka gas condensate fields. The effect of formation treatment with ammonium carbonate salts is achieved due to clearing of well bottom zone and increasing the formation permeability. At wells № 56, 108 of Yablunivka and № 58 of Tymofiivka gas condensate fields, an increase in the condensate ratio was observed by 22-35%. The effectiveness of this treatment is associated with the simultaneous bottomhole zone cleaning from asphalt-resinous contaminants and permeability increase, as well as with the hydrophilization of the pore space and mobility increase of condensate precipitated as a result of carbon dioxide effect, which was rejected as a result of decomposition of ammonium carbonate. Thus, experimental and industrial tests in Opishnia, Mashivka, Tymofiivka, Yablunivka gas condensate fields of Poltava region confirmed the effectiveness of using ammonium carbonate to increase hydrocarbon production. The prospect of further research is aimed at developing a technology for increasing the production of liquid hydrocarbons by using ammonium carbonate salts.

## Introduction

Ukraine has significant deposits of developed mineral deposits. To date, 90 types of minerals have been explored, which are concentrated in almost 8000 deposits. For sustainable development it is necessary to increase the efficiency of natural resources [1, 2, 3].

Today, most of the gas, gas condensate and oil fields in Ukraine are at the final stage of development, the production of gas and liquid hydrocarbons is decreasing.

One of the urgent problems of the oil and gas industry is to improve the efficiency of wells operation. In modern conditions, the solution to this problem is largely determined by the timely development, justification and targeted application of new technologies to improve the efficiency of hydrocarbon extraction.

The development of gas condensate fields for depletion is accompanied by a decrease in reservoir

pressure and retrograde condensation of high-boiling C<sub>5</sub>+ hydrocarbons (condensate) with the loss of some of them into the liquid phase and their loss in the reservoirs, due to the fact that the condensate that precipitates in the reservoirs turns out to be stationary and is not involved in filtration processes.

As a result, up to 30-60% of the initial condensate reserves remain in the deposits [4-6]. In addition, there is clogging of the pores with rock particles, asphalt, resin, paraffin deposits [7-9].

Therefore, the search and substantiation of new methods of field development aimed at increasing the condensate recovery of the productive formation, at the same time, will ensure the cleaning of the bottomhole zone from pollution products and this is an urgent task.

At the first stage of development of oil and gas condensate fields, production is carried out due to the depletion of reservoir energy. This stage, called primary,

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makes it possible to extract from the underground reservoir only up to 10-25% of hydrocarbons [10]. The various methods developed later were primarily aimed at maintaining reservoir pressure – methods of secondary production, or to improve the conditions for the movement of hydrocarbons in the porous environment of the rock – methods of tertiary production. The latter include various approaches to increasing the recovery of liquid hydrocarbons from depleted and highly watered fields.

These methods and their variations, according to some estimates [11], have up to 250 names, including various schemes for the formation of excess pressure, injection of working media, thermal effects on the formation, chemical treatment of productive reservoirs, and the like.

One of the most effective ways to increase the recovery of liquid hydrocarbons from reservoirs is to inject water and  $CO_2$  into the well [11-13]. With regard to the prospects for using the method of injecting carbon dioxide into the reservoir to increase oil and condensate recovery of depleted and highly watered fields, let us focus on the influence of the factor of carbon dioxide dissolution in water. Firstly, an aqueous solution of  $CO_2$ , entering into reactions and dissolving individual constituents of the rock, increases the permeability of the porous reservoir. Secondly, the viscosity of water increases, which leads to a decrease in its mobility, and together with an increase in the mobility of liquid hydrocarbons (through the dissolution of carbon dioxide in them), this contributes to the equalization of the mobility of water and liquid hydrocarbons, which is important for the efficiency of the process. Finally, changes in the properties of water and liquid hydrocarbons cause a decrease in the surface tension at «liquid hydrocarbons-water» interface and increase the wettability of the rock with water. All this in combination contributes to a more efficient washout of the hydrocarbon film. [14, 15].

According to 2000 estimates, there were 84 projects to enhance condensate and oil recovery by carbon dioxide injection in the world at the stage of implementation. Of these, 72 (96%) were carried out in the United States, which indicates its leadership in this important scientific and technological area. It is interesting that in work [16] the second place is given to Turkey, whose projects, according to expert estimates, allow counting on significant commercial success [16]. According to estimates, in 2014, 136 carbon dioxide injection projects are being implemented worldwide, carried out by 30 operators. Of these, 88 are considered successful, 18 are promising projects, the remaining 20 have recently started [17].

The use of carbonized water to enhance condensate and oil recovery is also known. The main advantage of the injection of carbonized water is the relatively low consumption of carbon dioxide during injection into the reservoir in comparison with other options for its use [18].

However, in both cases, pure liquefied carbon dioxide, a compressor, and special equipment for injecting liquefied gases and liquids saturated with gas at high pressures are required.

Based on the above, it can be assumed that a reagent containing 10-15% of chemically bound  $CO_2$ , which is release under formation conditions, can be highly effective.

Such a reagent is a salt of carbonate (carbonic) acid  $H_2CO_3$ , namely, bicarbonate and ammonium carbonate ( $NH_4HCO_3$  and  $(NH_4)_2CO_3$  respectively). Solutions of these salts decompose easily at temperatures of 40-90 °C. Therefore, in some cases, it is more reasonable from an economic point of view to use reagents containing 10-15% of chemically bound carbon dioxide  $CO_2$ , which is released in reservoir conditions.

The aim of the work is to study the effect of ammonium carbonate on the filtration properties of the reservoir, displacing and washing out the properties of the solution of ammonium carbonate and the use of a new reagent to increase condensate recovery at the fields of the Dnieper-Donetsk basin.

## Materials and methods

The object of the study is a 20% concentration of ammonium bicarbonate (ammonium carbonate) solution.

The influence of solutions of ammonium carbonate salts on the physical and lithological parameters of core samples, namely: permeability, porosity, size of pore channels, were studied at the UIPK-1M installation, for which the natural core was filled with ammonium carbonate salts at a concentration of 200 g/l at temperatures 30, 40, 50, 60, 70 °C for 3 hours.

Core material from various oil and gas fields (Khukhra, Reshetniaky, Rybalske, Velyki Bubny, Novohryhorivka and Matlakhivka) was used as reservoir models that were studied, namely, sandstone samples of 15 pcs. with the diameter of 30 mm and length 30 mm, with a permeability of 0.051-1.320  $\mu m^2$  and open porosity, which was in the range of 9.5-22.7%. Table 1 shows the geography of the studied samples.

Permeability was determined on a stationary laboratory setup with a fixed side hydraulic compression of 3 MPa.

Ammonium carbonate salts were removed from the core samples: in one case – with distilled water, in another – with formation water. The completeness of washing was determined by the presence of ammonium and carbonate ions in the filtrate by the titrimetric method.

The displacing properties of ammonium carbonate salts were determined by the value of phase permeability in the core samples. Phase permeability was investigated on the UIPK-1M installation at temperatures of 20, 30, 40, 50, 60 °C.

The washing properties of the solution of ammonium carbonate salts were investigated as follows: during the experiments, a sample of asphaltene substances (AS) dissolved in acetone, which were taken from the Yablunivka gas condensate field, was placed in the studied formation model. Acetone was removed by heating the test layer to 45 °C.

After that, the formation model was washed with a solution of ammonium carbonate with a volume concentration of 20%. After each washing, the formation

model was dried in a drying oven and the content of asphalt-resinous substances was determined by the gravimetric method. For comparison, another formation model was washed with formation water.

**Table 1.** Geography of the studied core samples.

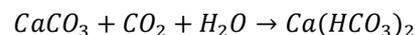
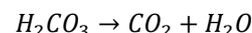
Core sample №	The field where core samples taken	Well №	Selection interval <i>h</i> m
4802	Khukhra	9	2931-2988
4805	Khukhra	9	2931-2988
4809	Khukhra	9	2931-2988
4814	Khukhra	9	3046-3053
4837	Novohryhorivka	100	3693-3697
4844	Novohryhorivka	100	3693-3697
4838	Novohryhorivka	100	3693-3697
4873	Matlakhivka	36	3577-3584
5021	Reshetniaky	65	2359-2367
5023	Reshetniaky	65	2359-2367
5027	Reshetniaky	65	2359-2367
4410	Velyki Bubny	114	2921-2929
4414	Velyki Bubny	114	2929-2936
4416	Velyki Bubny	114	2929-2936
5114	Rybalske	206	4284-4294

## Research results

The results of studying the effect of ammonium carbonate solutions on the physical and lithological parameters of core samples are shown in Table 2, 3.

Analysis of the data shows that the samples, from which ammonium carbonate salts were removed by distilled water, increased the permeability value by an average of 18% relative to the initial values. The theoretical size of the pore channels also increased in all studied core samples.

In addition, the effect of  $NH_4HCO_3$  solution on the carbonate content of the samples is observed. In almost all cores, it decreased by 47-52% from the initial values (Table 2). As a result of the experiments, it can be argued that the dissolution of carbonate cements with ammonium salts occurs according to the equation.



In this case, insoluble  $CaCO_3$  converted into soluble  $Ca(HCO_3)_2$ .

**Table 2.** Change of physical and lithological parameters of core samples during their treatment with  $NH_4HCO_3$  solution ( $t = 20^\circ C$ ) with subsequent removal of  $NH_4HCO_3$  with distilled water.

№ Core sample	Initial parameters			Final parameters			Carbonation <i>K</i> , %	
	Permeability <i>k</i> $10^{-15} m^2$	Porosity <i>n</i> %	Pore size <i>l</i> $\mu m$	Permeability <i>k</i> $10^{-15} m^2$	Porosity <i>n</i> %	Pore size <i>l</i> $\mu m$	Before	After
4802	940	20.8	19.0	1030	22.9	17.3	1	0.5
4805	1050	22.1	20.9	1326	22.6	18.4	3.3	1.6
4809	620	21.1	17.3	867	17.9	15.8	2.8	1.4
4814	42	14.2	5.4	57	11.8	5.0	3.1	1.6
4837	166	20.8	24.9	172	20.6	23.1	14	11
4844	46	9.9	6.9	64	9.6	5.9	9	7
5021	1292	22.7	20.4	1341	23.0	20.6	0.9	0.4

**Table 3.** Change of physical and lithological parameters of core samples during their treatment with  $NH_4HCO_3$  solution ( $t = 20^\circ C$ ) with subsequent removal of  $NH_4HCO_3$  by formation water.

№ Core sample	Initial parameters			Final parameters		
	Permeability <i>k</i> $10^{-15} m^2$	Porosity <i>n</i> %	Pore size <i>l</i> $\mu m$	Permeability <i>k</i> $10^{-15} m^2$	Porosity <i>n</i> %	Pore size <i>l</i> $\mu m$
4873	172	17.9	8.4	166	14.8	8.3
5027	337	21.7	10.6	310	21.3	10.2
4410	70	16.8	5.5	56	16.5	4.9
4416	245	22.4	8.9	192	17.6	8.9
4838	1038	21.8	18.6	890	18.5	18.6
5023	366	18.8	11.7	345	18.1	10.8
5114	51	10.4	6.0	40	10.2	5.3
5119	63	9.5	6.9	45	9.0	6.0
4414	290	22.6	9.7	246	22.2	8.9

In the laboratory, a number of experiments were performed to dissolve dry chalk with 20% ammonium bicarbonate solution.

The results showed that under normal conditions, the solubility of  $CaCO_3$  in  $NH_4HCO_3$  reaches 0.85-1.12 g/l, and at a temperature of  $60^\circ C$  – 1.6-1.74 g/l. This indicates

the solubility of carbonate cements, which contain a solution of ammonium bicarbonate in the core samples.

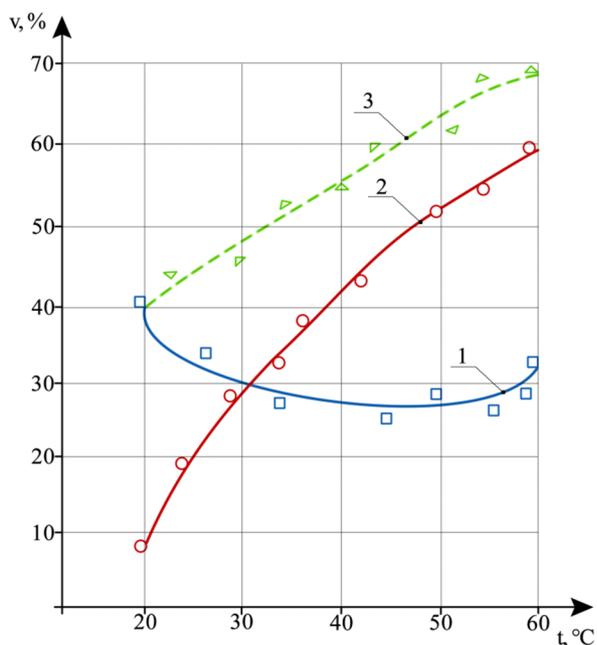
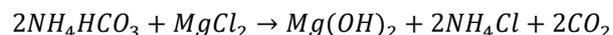
Based on the thermochemical conditions of this process, the solubility of carbonate cement will increase with increasing pressure, which is a positive characteristic

of the effect of  $NH_4HCO_3$  solution on carbonate rocks in formation conditions.

The second part of the core samples shown in Table 3, from which ammonium carbonate was removed by formation water, reduced the permeability value by 14-28%, the size of the pore channels also decreased.

The reason for this effect of ammonium bicarbonate solution on the permeability of the core samples is obviously a chemical reaction between highly mineralized water and ammonium bicarbonate with the formation and precipitation of a solid phase.

Laboratory studies carried out when mixing formation waters from gas condensate wells with a 20% ammonium bicarbonate solution showed that when the content of  $Ca^{2+}$  and  $Fe^{3+}$  in the formation waters is more than 0.5 mg-ion / l, as well as  $Mg^{2+}$  – 1.5 mg- ion/l and more, the use of ammonium carbonate is ineffective due to the formation of water-insoluble carbonates according to the scheme.



**Fig. 1.** Dependence of the relative permeability of core samples on temperature. Permeability: 1 – by solution of ammonium bicarbonate (in a sample of kerosene) 2 – by formation water (in a sample of kerosene), 3 – by air – (in a sample of kerosene).

Further research was aimed at studying the displacing properties of ammonium carbonate salts.

Table 4 shows the average values of the displacement coefficient, residual saturation, and relative permeability. From these data it follows that the relative permeability of the samples in descending order is as follows:

At a temperature of 20 °C:

1. by ammonium bicarbonate solution;
2. by air (in the kerosene sample);
3. by formation water (in the kerosene sample).

At a temperature of 60 °C:

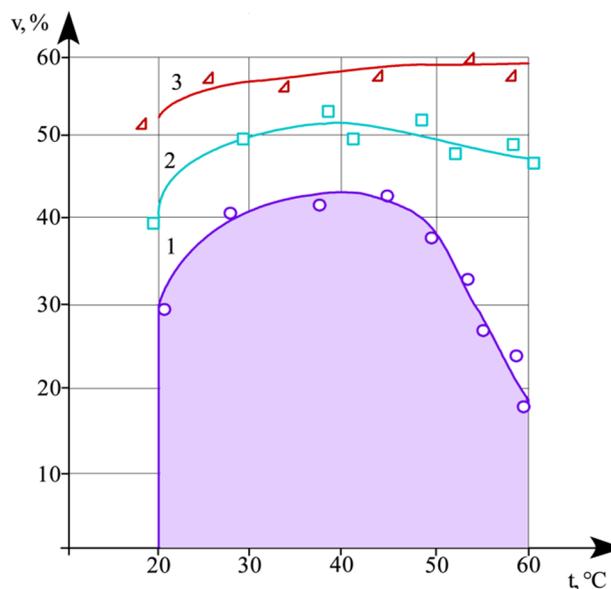
1. by air (in the kerosene sample);

2. by formation water (in the kerosene sample);

3. by ammonium bicarbonate (in the kerosene sample).

Based on the results of experiments for displacing kerosene from core samples, graphical dependencies were built, which are shown in Fig. 1 and 2.

The low phase permeability of the core with respect to ammonium bicarbonate confirms the well-known phenomenon of an increase in the viscosity of aqueous solutions when they are saturated with carbon dioxide. It is known [19] that with an increase in the concentration of  $CO_2$  in an aqueous solution up to 2.1%, the viscosity increases by 18.9%, and at a concentration of  $CO_2$  up to 5.6% – by 27.2%.



**Fig. 2.** Temperature dependence of the relative permeability of core samples. Displacement of kerosene from the core sample by: 1 – ammonium bicarbonate solution, 2 – formation water, 3 – air.

Analyzing the data shown in Fig. 2, it can be assumed that the increase in the temperature of the solution to 30-40 °C causes the release of some carbon dioxide, which initially pollutes the carbonate solution, increasing its viscosity, and at the same time blocks the pore space (Joumen effect). This, in turn, leads to a decrease in the phase permeability of the core to ammonium bicarbonate and low displacement of  $NH_4HCO_3$ .

However, a further increase in the temperature of the solution to 50-60 °C leads to an intensive release of carbon dioxide, which is dissolved in kerosene, changing its physicochemical properties. Studies show that as the temperature rises, the kerosene yield increases and the phase permeability of the degassed ammonia solution increases.

Further research is aimed at studying the washing properties of a solution of ammonium carbonate salts. During the experiments, a sample of asphaltene sediments (AS) dissolved in acetone, which were isolated from the Yablunivka gas condensate field, was placed in the studied formation model. Acetone was removed by heating the studied formation to 45 °C.

**Table 4.** The results of experiments to determine the coefficients of displacement, saturation and relative permeability.

№	Characteristics of the experiment	Parameters	The results of the experiment					
			Experiment temperature, °C					
			20	30	40	50	60	70
1.	Displacement of kerosene by ammonium bicarbonate solution	displacement coefficient, $\beta$	0.75	0.60	0.57	0.68	0.80	–
		saturation coefficient, $k_o$	0.25	0.40	0.43	0.32	0.20	–
		relative permeability, $k_i$	0.40	0.35	0.26	0.26	0.33	–
2.	Displacement of kerosene by formation water	displacement coefficient, $\beta$	0.60	0.50	0.48	0.50	0.55	0.60
		saturation coefficient, $k_o$	0.40	0.50	0.52	0.50	0.45	0.38
		relative permeability, $k_i$	0.11	0.28	0.42	0.54	0.58	0.62
3.	Displacement of kerosene by air	displacement coefficient, $\beta$	0.47	0.43	0.41	0.40	0.37	–
		saturation coefficient, $k_o$	0.53	0.57	0.59	0.60	0.63	–
		relative permeability, $k_i$	0.40	0.45	0.52	0.62	0.66	–

**Table 5.** Influence of washing on the content of asphaltene sediments in the formation model.

Temperature, °C	The content of asphaltene sediments, %					
	Formation water			20th solution of $NH_4HCO_3$		
	Washing number			Washing number		
	1	2	3	1	2	3
20	86	79	53	12	4	0.1
40	72	51	23	4.5	0.1	–
60	60	29	10	1.6	–	–
80	45	11	0.2	0.3	–	–

The formation model was then washed with a solution of ammonium carbonate salts with a volume concentration of 20%. After each washing, the formation model was dried in an oven and the content of asphaltene sediments was determined by weight. For comparison, washing of another model formation was performed with formation water. The research results are shown in Table 5.

As can be seen from the table, the experiments were performed at temperatures of 20, 40, 60, 80 °C, and the formation model was washed three times. Analysis of the research results shows that the formation water only after the third wash at a temperature of 80 °C allows cleaning the formation from asphaltene sediments. At lower temperatures, the washing efficiency is low.

The use of a 20% solution of ammonium carbon dioxide as a washing liquid shows that even at a temperature of 20 °C after a single wash in the formation remains only 12% of asphaltene compounds. The efficiency of formation treatment increases with increasing temperature. From 40 °C begins the release of carbon dioxide, which, in turn, leads to increased purification of the pore space. At a temperature of 80 °C, the model formation is cleaned practically with ammonia water and carbon dioxide, which are formed during the decomposition of ammonium carbon dioxide salts.

## The results of industrial tests

At the enterprises of GPD «PoltavaGasVydobuvannya» introduction of ammonium carbonate salts for treatment of the bottomhole zone of the formation was carried out. Investigations of wells after treatment were carried out on existing special research lines by the method of constant sampling in 5-6 modes of forward and 2-3 modes of reverse.

At the beginning of the test, the wells are flushed with 3-5% ammonia solution or ammonium bicarbonate, which are fed into the tubing or into the annulus. After that, the main intensification reagents are injected: ammonium carbonate salts.

The method of injection was chosen depending on the permeability of the collector. The working reagent was forced into the reservoir, kept there for 16-24 hours, then the well was developed in the barn and connected to the washing plant of complex gas treatment. After development and stabilization of the flow rate, the operating parameters of the well were removed.

The results of implementation showed that after treatment of productive reservoirs in wells № 23 of Opishnia and № 115 of Mashivka gas condensate fields (GCF) an increase in gas flow was obtained from 10 to 33.8 thous.m<sup>3</sup>/day and from 15 to 30.4 thousand m<sup>3</sup>/day,

and at well № 3 of Tymofiivka gas condensate field increase in gas production amounted to 80 thous. m<sup>3</sup>/day (Table 6).

Thus, the effect of bottomhole zone treatment of the formation with ammonium carbonate salts was achieved due to the cleaning of the bottomhole zone of the formation and permeability increase of reservoir.

Another effect of well intensification with  $NH_4HCO_3$  reagent is a significant increase in the condensate factor with some reduction in gas flow. Thus, at well № 56 of the Yablunivka gas condensate field, the condensate factor increased from 54.8 to 80.9 kg/thous.m<sup>3</sup>. The gas

productivity after intensification decreased from 180 to 130-140 kg/thous.m<sup>3</sup>. In the second month of well operation, after treatment of the reservoir, an increase in gas flow was observed, which is 55 thous. m<sup>3</sup>/day more than the initial values. Treatment of this well shows that the proposed reagent obviously increases the mobility of the condensate. The formation was hydrophilized with the displacement of liquid hydrocarbons from the pore channels, as evidenced by a sharp increase in condensate flow. After cleaning the reservoir, gas production was increased.

**Table 6.** Influence of intensification on well productivity.

Period	Gas flow rate $Q_g$ thous. m <sup>3</sup> /day	Condensate factor $CF$ kg/thous. m <sup>3</sup>	Condensate flow rate $Q_c$ , before treatment and calculation without taking into account $CF$ t/day
Before treatment	14.7	69.1	1.01
After treatment	95.14	69.1	6.6
1 month	90.6	69.1	6.2
2 month	50.6	69.1	3.5
3 month	15.0	69.1	1.03

**Table 7.** Influence of intensification on well productivity.

Period	Gas flow rate $Q_g$ thous.m <sup>3</sup> /day	Condensate factor $CF$ kg/thous. m <sup>3</sup>	Condensate flow rate $Q_c$ before treatment and calculation without taking into account $CF$ t/day	Condensate flow rate $Q_c$ after treatment t/day	Condensate flow rate $Q_c$ t/day
Tymofiivka GCF, well № 58					
Before treatment	80.0	101	8.1	–	–
After treatment	122.8	117	12.4	14.4	2.0
1 month	120.0	128	12.1	15.4	3.3
2 month	110.0	120	11.1	13.2	2.1
3 month	100.0	119	10.1	11.9	1.8
4 month	100.0	108	10.1	10.8	0.7
5 month	90.0	108	9.0	9.7	0.7
Yablunivka GCF, well № 108					
Before treatment	290	55	15.8	–	–
After treatment	200	71	10.9	14.2	3.3
1 month	322	80	17.6	25.8	8.2
2 month	316	74	17.3	23.5	6.2
3 month	314	70	17.1	22.3	5.2
4-6 months	310	68	16.8	20.9	4.7
7-9 months	307	60	16.0	18.6	2.6
10 months	290	55	15.9	–	–

The most efficient is the treatment of wells № 58 of Tymofiivka and № 108 of Yablunivka gas condensate fields. The data given in Table 7, indicate that in both wells after treatment increased gas flow rate and condensate factor. The gas flow rate gradually decreased to the initial values, while the condensate factor remains high.

The effectiveness of this treatment is obviously associated with the simultaneous cleaning of the bottomhole zone from asphalt-resinous contaminants and

an increase in permeability, as well as with the hydrophilization of the pore space and an increase in the mobility of condensate precipitated as a result of exposure to carbon dioxide, which was released as a result of decomposition of ammonium carbonate.

## Conclusions

1. Ammonium carbonate salts, in the absence of calcium chloride type waters, interact with carbonate rocks,

increase the permeability of reservoirs due to the conversion of insoluble  $CaCO_3$  to soluble  $Ca(HCO_3)_2$ .

2. Ammonium carbonate salts have high displacing properties, the coefficient of displacement of kerosene with  $NH_4HCO_3$  solution is 0.75-0.80, while reservoir water – 0.55-0.58.

3. The  $NH_4HCO_3$  solution has good washing properties against asphaltene contaminants. So, after washing the reservoir with ammonium carbonate salts at a temperature of 20 °C, no more than 10-12% of asphaltene substances remain in the model reservoir, and more than 60% with water.

4. Experimental and industrial tests at Opishnia, Mashivka, Tymofiivka, Yablunivka gas condensate fields of Poltava region confirmed the effectiveness of ammonium carbonate salts use for increasing hydrocarbon production. Industrial tests of ammonium carbonate salts showed an increase in gas flow by 30-50% at wells № 23 of Opishnia, № 115 of Mashivka, № 3 of Tymofiivka GCF, an increase in condensate factor by 22-35% at wells № 56 Yablunivka, № 58 of Tymofiivka GCR. The prospect of further research is aimed at developing a technology for increasing the production of liquid hydrocarbons by using ammonium carbonate salts.

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