



ISSN: 2350-0328

**International Journal of Advanced Research in Science,
Engineering and Technology**

Vol. 10, Issue 1, January 2023

Obtaining Complex Liquid Fertilizer NKS and NPKS Based on the Residual of a Nitrating Mixture Formed During the Nitration of 2-Methylquinazolin-4-one and Angren Brown Coal

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ABSTRACT: In this article, based on the mixture formed as a result of nitration reactions of 2-methylquinazolin-4-one, a solution of potassium humate obtained in the process of oxidation of brown coal of the Angren mine, obtained on the basis of oxidized coal products, amino acids, on the basis of ammonium sulfate, urea, potassium chloride and EPA (extraction phosphoric acid), an optimal method for obtaining complex humates NKS and NPKS has been developed, and the composition and properties of humates, saturated vapor pressure, density, viscosity and crystallization temperature have been studied and found to be interdependent.

KEYWORDS: heterocyclic compound, 2-methylquinazolin-4-one, nitration, brown coal, oxidized coal, humic acid, fulvic acids, potassium humate solution, amino acids, ammonium sulfate, urea, potassium chloride.

I. INTRODUCTION

Methods of synthesizing quinazolin-4-one, which is a heterocyclic compound, and its 2-substituted derivatives using anthranilic acid, formamide and its amides or anhydrides are presented in the literature [1-2]. In our previous research work, we have researched a method that is effective and convenient compared to the above methods, as well as a high-yield synthesis by using various catalysts, a systematic analysis of the obtained results based on chemical theories and laws, and bicyclic quinazolin-4-one and its 2 We have given information about the relative effectiveness of synthesis of -substituted compounds and methods [3].

This research paper presents the oxidation process of Angren lignite in the presence of the residue of the nitration mixture formed during the nitration reactions of 2-methylquinazolin-4-one in the presence of nitric and sulfuric acids, as well as the obtained results and their analysis.

II. SIGNIFICANCE OF THE SYSTEM

In this article, based on the mixture formed as a result of nitration reactions of 2-methylquinazolin-4-one, a solution of potassium humate obtained in the process of oxidation of brown coal of the Angren mine. The study of methodology is explained in section III, section IV covers the experimental results of the study, and section V discusses the future study and conclusion.

III. METHODOLOGY

The synthesis of quinazolin-4-one and its derivatives is presented in many literature sources, as a result of the nitration reaction of quinazolin-4-one and its homologues in the presence of sulfate and nitric acid (nitrating mixture), the corresponding 6-nitro compounds are obtained based on the following reaction equation can be synthesized [3]

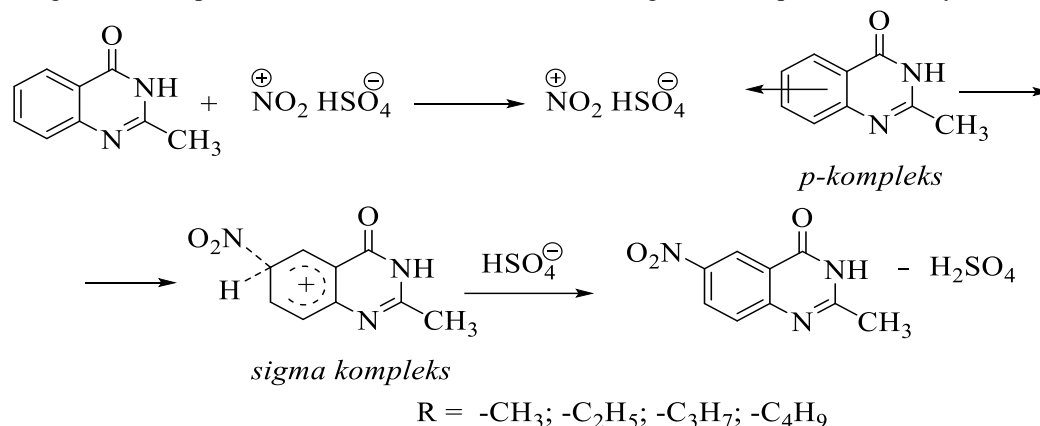


Figure 1. Nitration reaction of 2-R-quinazolin-4-one

Based on the nitration of compounds containing a benzene ring in the literature, taking into account that we can observe compounds containing two or more nitro groups, in order to introduce the second nitro group into the benzene ring of bicyclic quinazolin-4-ones, under different conditions and in the case of an increased amount of nitrating compound, the process is relatively tougher conditions, nitration reactions were carried out, but the formation of dinitrocompounds was not observed. This situation is explained by the reduction of the nucleophilicity of the benzene ring as a result of the effect of the amide carbonyl in the compound, the $-N=C$ bond and the nitro group included in the ring [3].

It should be noted that nowadays in agriculture nitrogen fertilizers, urea, ammonium nitrate, ammonium sulfate and potassium nitrate are well soluble in water and their efficiency is about 30-40% due to their low content of humus substances. does the rest washes into underground horizons and causes pollution of groundwater, reservoirs, lakes, rivers and seas. In addition, low efficiency leads to an increase in the demand for these fertilizers. When humus-containing mineral fertilizers are used, the humic substances contained in the fertilizers help to absorb nutrients and moisture from the soil, while reducing the possibility of nutrient leaching [4].

In the literature, it is proposed to mix equal amounts of brown coal and urea to obtain humic fertilizers [5]. The product is a free-flowing powder containing 23% nitrogen. The results of dissolution rate studies showed that nitrogen dissolves 8 times slower than urea from this fertilizer. Agrochemical tests conducted with barley showed that the fertilizer provides nitrogen nutrition to plants and has a stimulating effect on plant growth and development. The obtained fertilizers were also tested on vegetable crops. As a result of the use of this fertilizer, the yield of tomatoes increased by 18,2 cents/ha, carrots by 10.0 cents/ha, and the amount of nitrates in fruits decreased.

In the research conducted by experts in the field, the value and mechanism of action of humic substances and humic fertilizers in agricultural production are given [6-8]. Soluble humates are the most active substances in plant nutrition. They affect the general process of plant metabolism, increase oxygen absorption, activate enzymes, increase the formation of chlorophyll, change the forms of phosphorus compounds, increase mono- and disaccharides, as well as proteins in crops.

IV. EXPERIMENTAL RESULTS

In our study, a mixture of concentrated nitric and sulfuric acids was selected as a nitrating agent in the synthesis of 2-methyl-6-nitroquinazolin-4-one. Nitration reactions were carried out in a 500 mL four-necked flask equipped with an acid-resistant reflux condenser and a mechanical stirrer. Nitric acid ($\rho=1,65 \text{ g/cm}^3$) and concentrated sulfuric acid ($\rho=1,835 \text{ g/cm}^3$) were used as nitrifying mixture. The synthesis of 2-methyl-6-nitroquinazolin-4-one and the progress of the reaction process were carried out according to the methods presented in the literature [3, 9].

At this stage of the work, the process of obtaining liquid complex NCS and NPKS humates containing nitrogen, phosphorus, potassium, sulfur and soluble humic substances in various forms based on lignite oxidized with a nitrifying mixture using potassium hydroxide was studied. It should be noted that the selection of the proportions of the initial components was based on the fact that the obtained solutions can be used as plant growth stimulators. To obtain complex humates, we used a representative sample of fine coal from the Angren mine, which, after air-drying and pulverization in a ball mill to a size of 0,25 mm, had the following composition (wt.%): moisture 15, 26; ash 13,06; organic matter 71,68; GK 3,96 and fulvic acids 0,41, KCl (wt%) were used for organic mass. humidity – 5,2; Total K₂O – 60; ammonium sulfate

(% by weight) was used: moisture – 0,21; Total N – 21,1; SO₃ – 60; urea (wt.%): moisture – 0,3; Total N – 46,2 and purified neutralized extraction phosphoric acid (EFK).

The oxidation process is carried out for two hours at 40 °C with 5% HNO₃, and the organic mass ratio of coal, HNO₃:H₂SO₄=1:0,6:1,2 the coal obtained using it is processed. Oxidized coal produced by oxidation had the following composition: moisture – 1,58%, ash – 10,34%, organic matter – 88,08% and humic acids by organic mass of oxidized coal – 50,54%, fulvic acids – 2,06% and residual coal – 47,4%. To obtain humic acids from oxidized coal, it was treated with a 2,0 % solution of potassium hydroxide in the mass ratio of solid and liquid phases (Q:S): Q:S = 1:8. The extraction process was carried out in a mixer at a temperature of 80°C for 60 minutes, then the reaction mixture was separated into liquid and solid phases in a centrifuge, and the remaining solid phase was further processed in the second and third stages. Then, the solutions of the humates obtained in the third step were mixed and evaporated to a humidity of 95 % at a temperature not higher than 70 °C. Potassium chloride, amino acid and ammonium sulfate were added to the evaporated solutions of humate. The formed humate is mixed with potassium amino acids, ammonium sulfate, urea, potassium chloride and (extractable phosphoric acid) EFK until completely dissolved. As a result, NKS and NPKS humates with complex composition were obtained.

The density and viscosity of liquid fertilizers are determined in order to determine the conditions of storage, transportation and use of NKS and NPKS humates with a complex composition to increase soil fertility. The density was measured by the pycnometric method, and the viscometer was measured using a glass capillary viscometer with a diameter of 0.99 mm in the temperature range of 10-40 °C.

The obtained results are presented in Tables 1-4. As can be seen from the results presented in the tables, when a 5% solution of potassium humate is mixed with amino acids, ammonium sulfate, urea and potassium chloride, the amount of N is from 0,128 to 2,291%, potassium humate is from 4,504 to 5%, SO₃ is from 0,061 to 0,886%, AK is from 0,247 Complex NKS humates with 3,636 % and K₂O from 0,584 to 3,127% were obtained. By mixing potassium humate with 5% amino acids, ammonium sulfate, urea, potassium chloride and (extractable phosphoric acid) EFK, N content is from 0,248 to 2,361%, potassium humate is from 4,535 to 5%, SO₃ is from 0,056 to 0,296%, AK is from 0,246 to 3,636%, NPKS humates with K₂O from 0,580 to 2,582% and P₂O₅ from 0,061 to 0,955% were obtained.

Table 1

Chemical composition of liquid complex NKS humates obtained on the basis of potassium humate, potassium chloride, urea, amino acid and ammonium sulfate

Mass ratio Humate potassium: AK:KCl:CO(NH ₂) ₂ :(NH ₄) ₂ SO ₄	N, %	AK, %	K ₂ O, %	SO ₃ , %	Humate potassium, %	Moisture, %
100:0:0:0:0	0	0	0,44	0	5	95
A						
100:0,5:0,5:1:0,1	0,472	0,490	0,725	0,064	4,897	93,946
100:1:1:1:0,3	0,516	0,968	1,007	0,447	4,840	91,985
100:4:3:2:1	1,064	3,636	2,036	0,591	4,545	86,356
B						
100:0,5:0,5:1,5:0,1	0,250	0,487	0,846	0,063	4,873	92,593
100:1:1:1:0,3	0,516	0,968	1,007	0,447	4,840	91,985
100:2:5:2,5:0,5	1,159	1,818	3,127	0,295	4,535	86,344
D						
100:0,5:1:0,25:0,1	0,128	0,491	1,021	0,062	4,909	93,274

100:1:1:1,5:0,3	0,735	0,963	1,032	0,188	4,817	91,522
100:1,5:2,5:5:1	2,291	1,364	2,282	0,591	4,515	86,321
E						
100:0,25:0,25:0,5:0,1	0,251	0,247	0,584	0,061	4,946	93,166
100:1:1:1,5:0,5	0,774	0,962	1,002	0,313	4,808	91,346
100:2,5:3:3:1,5	1,655	2,273	2,032	0,886	4,504	86,302

Table 2
Flow viscosity of liquid complex NCS humates obtained on the basis of humate potassium, potassium chloride, urea, amino acid and ammonium sulfate

Mass ratio Humate potassium: AK : KCl : CO(NH ₂) ₂ : (NH ₄) ₂ SO ₄	Density (g/cm ³), temperatures, °C			Viscosity at temps (cPz), °C		
	10	20	40	10	20	40
100:0:0:0:0	1,034	1,015	1,006	1,344	1,132	0,918
A						
100:0,5:0,5:1:0,1	1,074	1,055	1,046	1,365	1,153	0,938
100:1:1:1:0,3	1,108	1,097	1,071	1,384	1,292	1,136
100:4:3:2,5:0,5	1,342	1,255	1,208	1,581	1,464	1,223
B						
100:0,5:1,5:0,5:0,1	1,054	1,031	1,016	1,714	1,386	1,018
100:1:1:1:0,3	1,108	1,097	1,071	1,384	1,292	1,136
100:2:5:2,5:0,5	1,364	1,242	1,228	1,914	1,592	1,356
D						
100:0,5:1:0,25:0,1	1,071	1,054	1,038	2,019	1,584	1,136
100:1:1:1,5:0,3	1,214	1,143	1,136	2,078	1,623	1,184
100:1:3:5:1	1,388	1,369	1,344	2,217	1,681	1,432
E						
100:0,25:0,25:0,5:0,1	1,081	1,063	1,057	2,052	1,614	1,221
100:1:1:1,5:0,5	1,273	1,176	1,177	2,163	1,719	1,322
100:2:4:3:1	1,454	1,427	1,407	2,343	2,083	1,625

Table 3
Chemical composition of liquid complex NPKS humates obtained on the basis of humate potassium, potassium chloride, urea, amino acid and ammonium sulfate and extractive phosphoric acid

Mass ratio Humate potassium: AK:KCl:CO(NH ₂) ₂ :(NH ₄) ₂ SO ₄ :EFK	N, %	AK, %	P ₂ O ₅ , %	K ₂ O, %	SO ₃ , %	Humate potassium, %	Moisture, %
100 : 0 : 0 : 0 : 0 : 0	0		0	0,44	0	5	95
A							
100 : 0,5 : 0,5 : 0,5 : 0,1 : 0,1	0,252	0,492	0,069	0,727	0,064	4,914	93,412
100 : 1 : 1 : 1 : 0,3 : 0,3	0,514	0,965	0,203	1,009	0,188	4,826	91,699

100 : 4 : 2 : 3 : 0,5 : 0,5	1,386	3,636	0,318	1,491	0,291	4,545	86,364
B							
100:0,5:0,25 : 0,5 : 0,1 : 0,1	0,250	0,490	0,067	0,582	0,062	4,929	93,642
100 : 1 : 1 : 1 : 0,3 : 0,3	0,514	0,965	0,203	1,009	0,188	4,826	91,699
100 : 2 : 4 : 1,25 : 0,25 : 0,5	0,589	1,818	0,316	2,582	0,148	4,535	86,346
D							
100:0,5 : 0,5 : 0,25 : 0,1 : 0,2	0,139	0,488	0,063	0,725	0,060	4,923	93,550
100 : 1 : 1 : 1 : 0,3 : 0,3	0,514	0,965	0,203	1,009	0,188	4,826	91,699
100:1,5:2,5:5,5 : 0,25 : 0,25	2,361	1,364	0,259	1,764	0,146	4,526	86,324
E							
100:0,25:0,5 : 0,5 : 0,1 : 0,1	0,248	0,246	0,061	0,721	0,058	4,920	93,638
100 : 1 : 1 : 1 : 0,5 : 0,3	0,554	0,958	0,196	1,003	0,180	4,811	91,522
100 : 2,5 : 1,5 : 4 : 1,5 : 0,5	1,982	2,273	0,314	1,218	0,296	4,522	86,360
F							
100:0,25 : 0,25 : 1 : 0,1 : 0,1	0,476	0,246	0,068	0,580	0,056	4,912	93,406
100 : 1 : 1 : 1 : 0,5 : 0,5	0,553	0,962	0,337	1,001	0,312	4,808	91,346
100 : 3 : 3 : 2 : 0,5 : 1,5	0,841	2,727	0,955	2,036	0,288	4,540	86,358

Table 4

Humate potassium, potassium chloride, urea, amino acid and ammonium sulfate and extractive phosphoric acid based liquid complex NPKS humate flow viscosity

Mass ratio Humate potassium: AK:KCl:CO(NH ₂) ₂ :(NH ₄) ₂ SO ₄ :EFK	Density (g/cm ³), temperatures, °C			Viscosity at temps (cPz), °C		
	10	20	40	10	20	40
100 : 0 : 0 : 0 : 0 : 0	1,034	1,015	1,006	1,344	1,132	0,918
A						
100 : 0,5 : 0,5 : 0,5 : 0,1 : 0,1	1,112	1,093	1,074	1,478	1,216	1,079
100 : 1 : 1 : 1 : 0,3 : 0,3	1,174	1,145	1,123	1,526	1,254	1,121
100 : 4 : 2 : 3 : 0,5 : 0,5	1,312	1,293	1,271	1,762	1,413	1,274
B						
100 : 0,5 : 0,25 : 0,5 : 0,1 : 0,1	1,131	1,110	1,093	1,745	1,388	1,118
100 : 1 : 1 : 1 : 0,3 : 0,3	1,174	1,145	1,123	1,526	1,254	1,121
100 : 2 : 4 : 1,25 : 0,25 : 0,5	1,394	1,358	1,335	2,088	1,712	1,435
D						
100 : 0,5 : 0,5 : 0,25 : 0,1 : 0,2	1,152	1,135	1,142	2,122	1,694	1,324
100 : 1 : 1 : 1 : 0,3 : 0,3	1,174	1,145	1,123	1,526	1,254	1,121
100 : 1,5 : 2,5 : 5,5 : 0,25 : 0,25	1,564	1,511	1,463	2,482	2,137	1,736
E						
100 : 0,25 : 0,5 : 0,5 : 0,1 : 0,1	1,162	1,144	1,128	2,424	1,853	1,432



100 : 1 : 1 : 1 : 0,5 : 0,3	1,255	1,216	1,164	2,542	1,988	1,523
100 : 2,5 : 1,5 : 4 : 1,5 : 0,5	1,636	1,582	1,523	2,781	2,356	2,043
F						
100 : 0,25 : 0,25 : 1 : 0,1 : 0,1	1,199	1,143	1,108	2,596	1,916	1,589
100 : 1 : 1 : 1 : 0,5 : 0,5	1,383	1,346	1,284	2,721	2,111	1,642
100 : 3 : 3 : 2 : 0,5 : 1,5	1,728	1,651	1,583	2,796	2,169	1,689

As can be seen from the tables, the density of solutions increases with increasing concentration of solutions and decreasing temperature. In the temperature range of 10-40 °C pulp viscosity and proportions humate : amino acids : potassium chloride : urea : ammonium sulfate : EFK = 100 : (0.25-4):(0.25-5) : (0.25-5 .5):(0.1-1.5) : (0.1-1) has low and high values. This means that the solution is transportable and has no difficulty in packaging it from device to device and then using it. At the same time, when humate : amino acids : potassium chloride : urea : ammonium sulfate : EFK = 100:1:1:1:0.3:0.3, due to the increase in the concentration of the solution, the formation of precipitation increases with the ratio humate : amino acids : potassium chloride : urea : ammonium sulfate : EFK = 100:(2-4):(2-5):(2-5.5):(0.5-1.5):(0.5- 1.5).

Vapor pressure of liquid complex NPK and NPKS humates was determined by dynamic method. It is known that in order to reduce the pressure of the gas phase on the solution, it is necessary to add a soluble gas to the solution. The temperature dependence of the change of the saturated vapor pressure in relation to the solutions of the liquid complex humates of NKS and NPKS obeys the $\lg P=A-B/T$ equation. Using the method of least squares, the values of constants A and B were calculated, and empirical equations were obtained for determining the vapor pressure of liquid fertilizers at other temperatures. Depending on the concentration of NPK and NPKS humate solutions, the values of A and B vary between 6.1095-4.9475 and 942.3-606.3, respectively. In the temperature range of 20-50 °C, the saturated vapor pressure of NKS and NPKS humates is 11.77-25.52 kPa, which indicates their low volatility even at high temperatures.

Table 5
Saturated vapor pressure (kPa) of solutions of liquid complex NKS humates obtained on the basis of potassium humate, potassium chloride, urea, amino acid and ammonium sulfate

Mass ratio Humate potassium: AK:KCl:CO(NH ₂) ₂ : (NH ₄) ₂ SO ₄	The form of the equation $\lg P=A-B/T$	Moisture, %	Temperatures, °C			
			20	30	40	50
100: 0 : 0 : 0 : 0	$\lg P=6,1095-938,3/T$	95	8,12	10,84	13,41	17,93
A						
100 : 0,5 : 0,5 : 1 : 0,1	$\lg P=5,6195-1142,3/T$	93,946	11,34	15,26	20,63	26,49
100 : 1 : 1 : 1 : 0,3	$\lg P=5,4475-1016,3/T$	91,985	12,76	16,71	21,96	27,98
100 : 4: 3 : 2,5 : 0,5	$\lg P=4,9585-1092,3/T$	86,356	16,88	20,82	26,04	32,04
B						
100:0,5 : 1,5 : 0,5 : 0,1	$\lg P=5,6145-1101,3/T$	92,593	11,12	15,03	20,41	26,24
100 : 1 : 1 : 1 : 0,3	$\lg P=5,4435-982,3/T$	91,945	12,25	16,49	21,74	27,71
100 : 2 : 5 : 2,5 : 0,5	$\lg P=4,9545-1053,3/T$	86,344	16,64	20,61	25,83	31,82
D						
100 : 0,5 : 1 : 0,25 : 0,1	$\lg P=5,6105-1065,3/T$	93,274	10,91	14,86	20,22	26,04
100 : 1 : 1 : 1,5 : 0,3	$\lg P=5,4392-1044,3/T$	91,522	11,84	16,34	21,52	27,49
100 : 1 : 3 : 5 : 1	$\lg P=4,9585-1017,3/T$	86,321	16,43	20,33	25,46	31,54
E						
100:0,25:0,25:0,5 : 0,1	$\lg P=5,5892-1038,3/T$	93,166	10,53	14,46	19,83	25,59
100 : 1 : 1 : 1,5 : 0,5	$\lg P=5,4545-978,3/T$	91,346	11,86	18,91	21,06	27,11
100 : 2 : 4 : 3 : 1	$\lg P=4,9475-904,3/T$	86,302	16,01	20,02	25,14	31,14

Table 6

Saturated vapor pressure (kPa) of liquid complex NPKS solutions of humates obtained on the basis of humate potassium, potassium chloride, urea, amino acid and ammonium sulfate and extraction phosphoric acid

Mass ratio Humate potassium: AK:KCl:CO(NH ₂) ₂ : (NH ₄) ₂ SO ₄ : EFK	The form of the equation lgP=A-B/T	Moisture, %	Temperatures, °C			
			20	30	40	50
100: 0 : 0: 0: 0	lgP=6,1095-938,3/T	95	8,12	10,84	13,41	17,93
A						
100:0,5:0,5:0,5:0,1:0,1	lgP=5,5195-942,3/T	93,412	12,36	17,26	22,73	27,73
100:1:1:1:0,3:0,3	lgP=5,3465-868,3/T	91,699	13,78	18,71	23,04	29,22
100:4:2:3:0,5:0,5	lgP=5,1785-836,3/T	86,364	15,06	19,22	24,45	30,69
B						
100:0,5:0,25:0,5:0,1:0,1	lgP=5,5134-938,3/T	93,642	12,04	16,96	22,42	27,42
100:1:1:1:0,3: 0,3	lgP=5,3431-828,3/T	91,684	13,43	18,41	22,74	28,82
100:2:4:1,25:0,25:0,5	lgP=5,1733-716,3/T	86,346	14,72	18,92	24,12	30,28
D						
100:0,5:0,5:0,25:0,1:0,2	lgP=5,5124-918,3/T	93,550	11,92	16,86	22,32	27,32
100:1:1:1:0,3:0,3	lgP=5,3392-798,3/T	91,682	13,34	18,31	22,64	28,68
100:1,5:2,5:5:5:0,25:0,25	lgP=5,1705-706,3/T	86,324	14,64	18,72	24,03	30,21
E						
100:0,25:0,5:0,5:0,1:0,1	lgP=5,5111-888,3/T	93,638	11,84	16,73	22,21	27,23
100:1:1:1:0,5:0,3	lgP=5,3382-788,3/T	91,522	13,18	18,22	22,44	28,57
100:2:1,5:5:1:0,5	lgP=5,1705-676,3/T	86,360	14,53	18,62	23,91	30,14
F						
100:0,25:0,25:1:0,1:0,1	lgP=5,5101-848,3/T	93,406	11,76	16,66	22,11	27,03
100:1:1:1:0,5:0,5	lgP=5,367-748,3/T	91,346	13,04	18,01	22,54	28,42
100:2:5:1,5:0,5:1	lgP=5,1692-606,3/T	86,358	14,33	18,48	23,55	29,89

The crystallization temperature of NKS and NPKS humates was determined by the visual-polythermal method. It varies from (- 5,5) to 28,0 °C, which allows them to be widely used as liquid plant stimulants and fertilizers at any time of the year.

V. CONCLUSION AND FUTURE WORK

The following can be concluded from the above studies.

Optimum conditions for obtaining complex humates of NKS and NPKS on the basis of humate obtained on the basis of oxidized coal products, amino acids, ammonium sulfate, urea, potassium chloride and (extraction phosphoric acid) EFK were studied.

From the point of view of agrochemistry, relative to the initial components to obtain NKS humate, humate : amino acids : potassium chloride : urea : ammonium sulfate = 100:1:1:1:0,3 which is N content 0,516 %, potassium humate 4,840 %, SO₃ 0,447 %, AK 0,968 % and K₂O 1,007 % and the ratio of initial components to obtain NPKS humate is humate : amino acids : potassium chloride : urea : ammonium sulfate : EFK= 100 : 1 : 1 : 1 : 0.3 : 0.3 is the amount of N 0.514%, potassium humate 4,826%, SO₃ 0,188 %, AK 0,965 %, K₂O 1,009 % and P₂O₅ 0,209 % have been found to have a suitable content as a plant growth stimulator.

The results of the conducted studies show that all types of developed NKS and NPKS humates have satisfactory physico-chemical properties that ensure their stability under long-term storage conditions.

Thus, the results of research reliably show the possibility of obtaining fertilizers containing various liquid soluble humus stimulants and various forms of nitrogen, soluble forms of humus substances, as well as sulfur.



ISSN: 2350-0328

International Journal of Advanced Research in Science, Engineering and Technology

Vol. 10, Issue 1, January 2023

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