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OPTIMIZATION OF THE ANALYTICAL REAGENT (AR) $Co_3(PO_4)_2 \cdot 8H_2O$ SYNTHESIS CONDITIONS USING THE HARRINGTON'S DESIRABILITY FUNCTION

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The Box-Wilson experiment design for the acceleration and optimization of the analytical reagent (AR) $Co_3(PO_4)_2 \cdot 8H_2O$ synthesis conditions was made. As optimization criteria of the three callback functions the Harrington's desirability function, which unites the different (of physical content) requirements to the examining object and conditions of its preparation was used. On the stage of replica executing of Box-Wilson Central Composite Design (CDD) was the optimal conditions completely corresponded to the specified requirements were determined. So, it wasn't need to design the polynomial model and finding local condition for this multivariable function. The correspondence degree of the chemical composition of isolated salt to the requirements "chemically pure" chemical (AR) by chemical analysis and X-ray method was demonstrated.

Introduction. The traditional methods of distinguishing the reasons for the synthesis of the chemical substances are connected with different experiments that take a lot of time and effort as they are mostly the 'passive' ones, which means that they are based on the varying of separate independent changeable figures in the conditions when the rest are being measured by the unchangeable or constant figures. These experiments are mainly multivariate and connected with the simultaneous optimization of several parameters. Thus, regardless of a great majority of performed synthesis, many decisions are being taken on the bases of casual information and are far from being perfect because it's impossible to investigate all possible combinations of influence factors. A nice way out can be statistical methods of experiment planning that were proposed by R. Fisher who proved the importance of the simultaneous variation of all the influence factors in comparison with the widespread single-factor experiment [1]. The next stage - is the planning of the extreme experiment that was first proposed by Box and Wilson [2]. Its main idea lay in the consecutive performance of small series of researches where all the factors simultaneously vary according to certain rules. The series are organized in such a way that after the mathematical conversion of the previous one it is possible to plan the performance conditions of the next series up to the achievement of the field of optimization. One of the great priorities of the method is its universal nature, its suitability for the search of optimal figures of different technical processes, for instance, in the food,

Table 1. The basic meanings of Harrington's psycho-physiological desirability function [10].

The quantitative mark on the Harrington's desirability scale	d(Y) criterion desirability meaning
0,80 – 1,00	Very good
0,63 – 0,80	Good
0,37 – 0,63	Satisfactorily
0,20 – 0,37	Badly
0,00 – 0,20	Very badly

microbiological, agricultural and biotechnical industries [4–7] and even in politics [9]. The useful way of consolidation of different physical criteria of optimization in one measurement is the integrated Harrington's desirability function [10]. It emerged in the process of investigation of the real decisions being taken by experimentalist and it has such useful features as integrity, monotony and coherence. The mathematical device of recalculation of specific parameters into abstract numerical values uses the logical function of E. K. Harrington – d – the so called 'desirability curve' that defines the empirically created function with two spaces of saturation (in $d \to 0$ and $d \to 1$) as well as linier space (from $d = 0{,}20$ up to $d = 0{,}63$). The Y – axis is the scale of frequency rates, and d – axis is the scale of desirability. The interval of effective meanings on the scale of frequency factors lies between the rates from -6 up to +6, though other variants are possible. Table 1 shows the rules of desirability scale building [10].

Such choice of numbers is explained by the convenience of calculation, as $d = 0{,}6 \sim 1 - 1/e$, and $d = 0{,}37 \sim 1/e$.

The d-scale is non-dimensional so with its help any reply can be transferred in such a way that it could be interpreted in terms of usefulness and desirability in any field - from chemical synthesis to political science, medicine, ecology [11] and even the evaluation of production compatibility.

Results and discussion. Synthesis of $Co_3(PO_4)_2 \cdot 8H_2O$ was performed in the following way: a fixed volume of distilled water was being heated up to the desired temperature. While stirring the mixture it was added the H_3PO_4 solution of the certain concentration till pH 3,0–3,3 was reached, as well as the solid powdered cobalt(II) hydroxocarbonate. The reagents being mixed they continued the synthesis during the given duration of synthesis. The volume of the mixture was kept constant by the adding of distilled water, heated to the temperature of synthesis. After finishing the process the suspension was cooled, the sediment was taken away in the process of filtration made in the vacuum filter, was washed in distilled water and dried to the constant weight at 40-60°C.

The CoO content in the products of synthesis was measured by the chelatometry [13]; P_2O_5 – by the gravimetric quinoline-molybdenum method [14]; H_2O – gravimetrically [15]; CO_2 – by means of volume gasometry [16]. The supernatant liquid was collected and measured as for its P_2O_5 content.

Development of a generalized parameter of the $Co_3(PO_4)_2 \cdot 8H_2O$ synthesis optimization. When providing the mathematical modeling of the process of obtaining the hydrated cobalt (II) orthophosphate three factors are being taken into consideration that have a direct affect on the quality of the final product and its output. As the requirements for $Co_3(PO_4)_2 \cdot 8H_2O$ are based on its purpose – the AR reagent – shouldn't contain more than 0.1% of impurities according to the requirements of DSTU (State

Standard of Ukraine 2216-93. Reagents and supra pure substances. Designations and methods for determination of purity degrees. Terms and definitions), and environmental requirements and efficiency – the growth solution should contain fewer residues of initial reagents. So, factors for optimization include:

1. Molar ratio of K=CoO:P_2O_5 should be as close as possible to 3, which corresponds its theoretical value in the individual compound – Y_1;

2. The content of impurities CO_2 (% wt.) is supposed to be minimal (the requirements for reactive compound purity should be not more than 0.1%) – Y_2;

3. The yield of the final product (%) for P_2O_5 should approach 100% or filtrate should contain the minimal content of CoO – Y_3. Indirectly, this factor also characterizes the efficiency of the process.

Schematic representation of the empirical process model of the "black box" is shown in Fig. 1. This term is called a non-transparent "box" in the system – oriented approach, which is separated from the environment, but is nevertheless associated with it due to inputs and outputs. It is assumed that the internal content of the system is unknown and its exploration has no meaning or purpose. The model in this case reflects two important and essential features of the system. First of all, it's integrity and isolation from the environment. Secondly, it doesn't exclude the influence of unaccounted factors also called disturbing factors which are unknown to the researcher. A special procedure is performed to clarify the issue of reproducibility tests results which are accountable to the response function for constancy control factors by conducting parallel experiments.

No factor (Y_1, Y_2, Y_3) can be used separately as the sole criterion for optimization of the synthesis process, as this will be considered only one-way requirements for the object of study. To optimize the process it was necessary to combine performance with different physical meaning and dimension. In order to obtain the generic factor that will satisfy the demands we used the Harrington's desirability function [10]. In the basis of this method of generalization lies the transformation of natural values of particular factors (y_1, y_2, y_3) into dimensionless quantities (in this system d_1, d_2 and d_3) with their subsequent combination according to the following formula:

$$Dgen.=\sqrt[n]{\prod_{i=1}^{n} d_i}. \qquad (1)$$

The following relationship is used to determine the value of di, which corre-

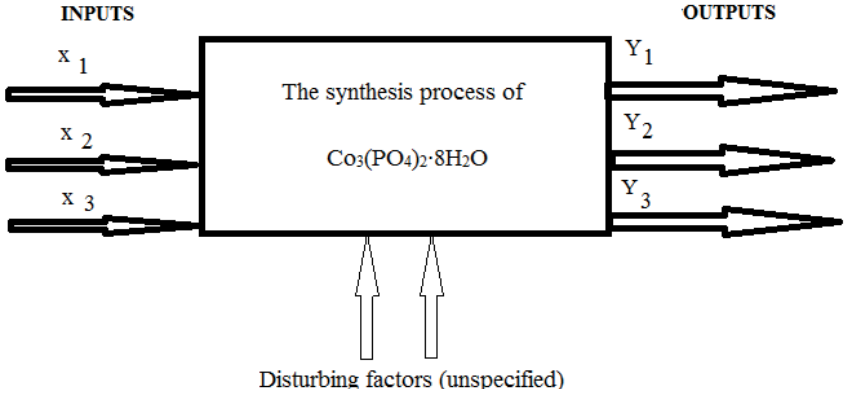

Fig. 1. Mathematical model of synthesis

Table 2. Example of the B_{ok} and B_{1k} coefficients calculation for the one-sided (Y_2) and two-sided limitation (Y_1)

The number of k interval	The molar ratio K = $CoO:P_2O_5$ - Y_1			The content of CO_2 impurities (% wt.) - Y_2		
	Physical values of intervals boundaries	B_{ok}	B_{1k}	Physical values of intervals boundaries	B_{ok}	B_{1k}
1 (very good)	2,97 – 3,03	-54,282	18,0941	0,10 - 0	-6,00	45,00
2 (good)	2,95 – 2,97 3,03 – 3,05	-28,3607 -27,8572	9,3663 9,3663	0,20 – 0,10	-2,23	7,28
3 (satisfactorily)	2,90 – 2,95 3,05 – 3,10	-15,3086 -14,9628	5,1452 5,1452	0,30 – 0,20	-2,32	7,72
4 (badly)	2,90 – 2,80 3,10 – 3,20	- 7,5558 - 6,0339	2,2650 2,2650	0,50 – 0,30	-0,75	2,50
5 (very badly)	2,70 - 2,80 3,20 – 3,30	-51,2245 -55,9448	17,8609 17,8609	1,00 – 0,50	-5,00	11,00

sponds y1, that was obtained in the process of chemical analysis of the sample of $Co_3(PO_4)_2 \cdot 8H_2O$ and the growth solution:

$$d = \exp[-\exp(-y')], \quad (2)$$

$$d = \exp[-\exp(y')], \quad (3)$$

where $y' = -6 \div +6$, that corresponds to the boundary values of Dgen. [10].

The di value can be determined graphically, but because of the low accuracy of this method we used the analytical method for the determination of d_i, and then — Dgen. Since it is assumed that within certain intervals of y' the spaces of Y_i — scale are uniform, the transition from Y_i to y' was performed with the help of the first order polynomial:

$$y' = B_{ok} + B_{1k} \cdot Yi, \quad (4)$$

where k — is the number of the interval containing the value of Y_i.

The numeric values of the Bok and B_{1k}

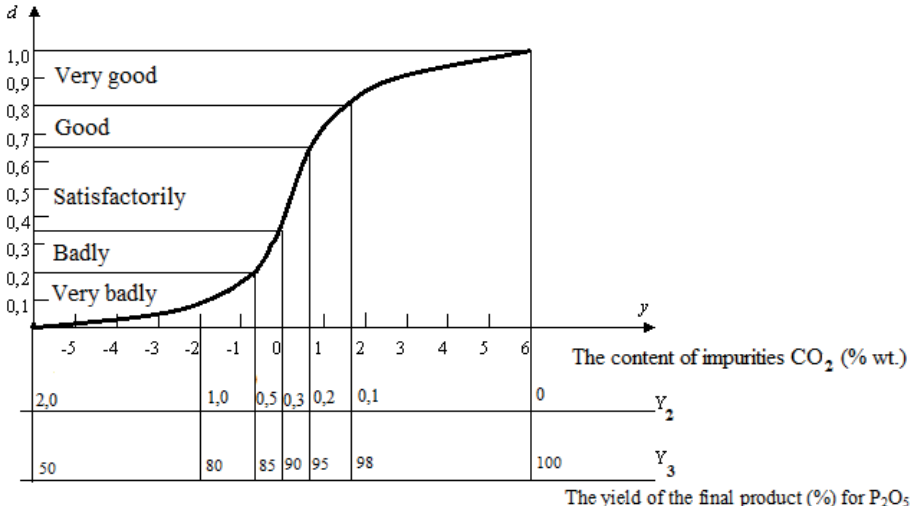

Fig. 2. Harrington's desirability function d=f(y′) used for the factors with one-sided limitation (Y_2 та Y_3)

coefficients were determined from the system of two equations for each interval (example shown in Table 2).

Figures 2 and 3 present the charts of d=f(y′) for one-sided and two-sided limitation variants. Scales Y_1, Y_2, Y_3 are also shown in these charts as the physical units with the qualitative assessment of their borders.

The scales cover the results of the previous investigations as well as demands for the $Co_3(PO_4)_2 \cdot 8H_2O$ quality and the process of its preparation.

The following equation is used for reaching di in the case of two-sided limitation of the Yi meaning:

$$d = \exp[-(y')^{-n}], \qquad (5)$$

where n – is the added number ($0 < n < \infty$).

The n index can be calculated by giving d meaning to some abstract quantity y (giving preference to the interval $0{,}6 < d < 0{,}9$) according to the formula (5):

$$n = \frac{\ln \ln \frac{1}{d}}{\ln |y'|}, \qquad (6)$$

and y′ is being calculated according to the formula (7):

$$y' = \frac{2Y - (Y_{max} + Y_{min})}{Y_{max} - Y_{min}} \qquad (7).$$

In the case which is discussed in this research the two-sided restriction refers to the criteria of optimization Y_1 – molar ratio $K=CoO:P_2O_5$. The chemical nature of this phenomenon is connected with the fact that in the target orthophosphate

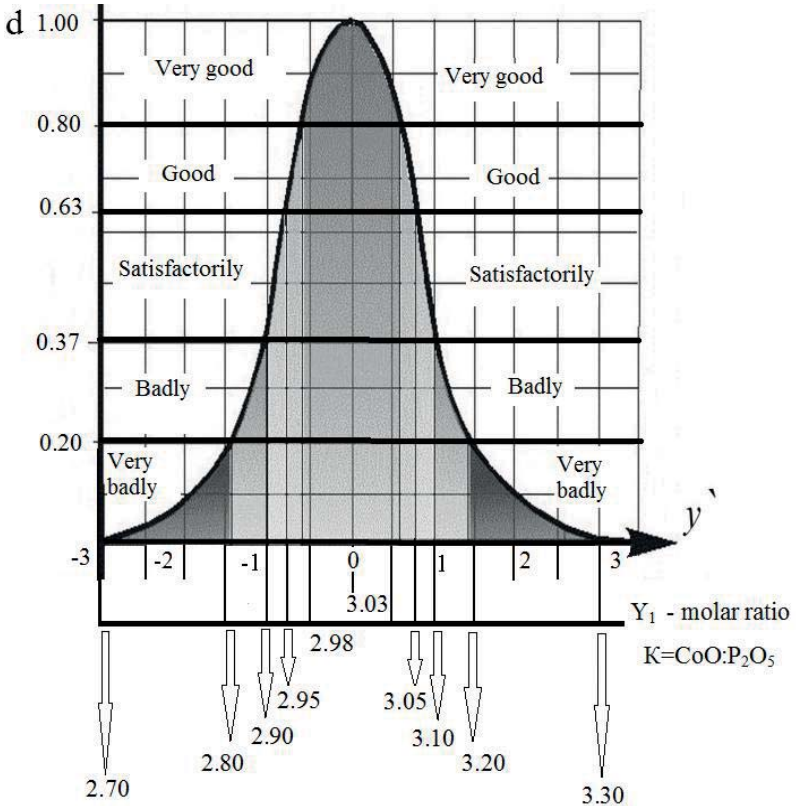

Fig. 3. Harrington's desirability function d=f(y′) for the two-sided factor (Y_1)

Table 3. Estimation of the experimental unit and results reproducibility

№	Results of experiments			$Y_1 - \bar{Y}_1$	$Y_2 - \bar{Y}_2$	$Y_3 - \bar{Y}_3$
	The composition of products of a synthesis		The yield of the final product (%) за P_2O_5 – Y_3			
	Molar ratio K= $CoO:P_2O_5$ – Y_1	The content of impurities CO_2 (% wt.) – Y_2				
1	2,97	0,10	95,19	-0,0125	-0,015	0,10
2	2,99	0,13	95,27	0,0075	0,015	0,18
3	2,98	0,11	94,97	-0,0025	-0,005	-0,12
4	2,99	0,12	94,93	0,0075	0,005	-0,16

Continuation of table 2

№	$(Y_1 - \bar{Y}_1)^2$	$(Y_2 - \bar{Y}_2)^2$	$(Y_3 - \bar{Y}_3)^2$	d_1	d_2	d_3	$D_{gen.}$
1	0,000156	0,000225	0,0100	0,8000	0,8000	0,9666	0,6186
2	0,000056	0,000225	0,0324	0,9851	0,7576	0,9684	0,7227
3	0,000006	0,000025	0,0144	0,9209	0,7866	0,9612	0,6963
4	0,000056	0,000025	0,0256	0,9851	0,7725	0,9601	0,7306
∑	0,000274	0,000500	0,0824				

The end of table 2

№	$\bar{D}$ gen	$D_{gen.} - \bar{D}_{gen.}$	$(D_{gen.} - \bar{D}_{gen.})^2$	$\sum (D_{gen.} - \bar{D}_{gen.})$	$\sum (D_{gen.} - \bar{D}_{gen.})^2$
1	0,6921	-0,07345	0,0053950	-0,00015	0,0078313

there can be present different compounds as unwanted impurities such as hydro phosphate composition $CoHPO_4$, in which the molar ratio is $K=CoO:P_2O_5 = 2$, and hydroxyl phosphate of the composition is Co_2OHPO_4, which is $K=CoO:P_2O_5 = 4$. It is possible to anticipate that this criterion of optimization will depend on the amount of H_3PO_4 in the synthesis. When it in excess the impurity of acid salt $CoHPO_4$ is possible, and when it lacks the impurity of basic salt Co_2OHPO_4 is obvious.

Before studying the dependence of the generalized criterion of optimization on factors that determine the preparative technology $Co_3(PO_4)_2 \cdot 8H_2O$, it is necessary to estimate the reproducibility of synthesis results and quality of an experimental unit.

The estimation of reproducibility of experimental results and quality of a laboratory unit. To estimate the reproducibility of the results of experimental researches four parallel experiments were done, the results of which are given in Table 3. The conditions of experiments met the centre of the plan of a partial replica according to

Table 4. The FFE matrix

№	Conditions of experiments	x_1, temperature, ⁰C	x_2, concentration H_3PO_4, % (wt.)	x_3, synthesis duration, hours.
1	Basic level (x_i^0) – plan centre	50	30	2
2	The period of variation (Δx_i)	30	20	1
3	Upper level ($x_{i\,max}$)	80	50	3
4	Lower level ($x_{i\,min}$)	20	10	1

DCC Box-Wilson: the temperature is 50°C; the H_3PO_4 solution concentration 30 (% wt.); the synthesis duration is 2 hours.

The reproducibility error was calculated according to the dispersion of parallel results formula:

$$S_{Yi} = \sqrt{\frac{\sum_{n=1}^{N}(Y_{n_i} - \bar{Y}_i)^2}{N-1}}, \text{ де} \qquad (8)$$

i – the number of a response function;

N– the number of parallel experiments.

Using the received data for calculation according to the formula (8), the following dispersion data SYi are received:

S_{Y1}=0,009557; S_{Y2}=0,0129073;

S_{Y3} = 0,1657302.

The calculation of the variation coefficient Vi is done using the formula:

$$V_i = \frac{S_{Yi} \cdot 100}{\bar{Y}_i}, \% \qquad (9)$$

Then

$$V_1 = \frac{0{,}009557 \cdot 100}{2{,}9825} = 0{,}31\%,$$

$$V_2 = \frac{0{,}0129073 \cdot 100}{0{,}115} = 11{,}22\%,$$

$$V_3 = \frac{0{,}1657302 \cdot 100}{95{,}09} = 0{,}17\%.$$

When the consideration of response function Y_1 and Y_3 meaning of the variation coefficient do not exceed 3%, it shows a good reproducibility of results. When considering the response function Y_2 (the CO_2 content in products) the coefficient of variation is 11,22%, it is connected with high relative error of CO_2 content definition using the volume gasometry method, which is 10% [16]. So the means of a synthesis unit be relevantly used to receive $Co_3(PO_4)_2 \cdot 8H_2O$.

Similarly to the described way above the dispersion of Harrington's desirability function was estimated using the formulae:

$$V = \frac{S_{D_{общ.}} \cdot 100}{\bar{D}_{об.}}, \% \qquad (10)$$

Table 4. The conditions results of experiments according DCC Box-Wilson plan of type 2^k, are fulfilled for the optimization of the process for getting $Co_3(PO_4)_2 \cdot 8H_2O$

№ experiment	Conditions of experiments – managing factors				The results of experiments								
	physical x_i		conditional X_i		Molar ratio K= $CoO:P_2O_5$ – Y_1	The content of impurities CO_2 (%) – Y_2	The yield of final product (%) for P_2O_5 – Y_3	Partial desirability, d_i			D_{gen}
	x_1, temperature, C	x_2, concentration H_3PO_4, % (wt.)	x_3, synthesis duration, hours	X_1	X_2	X_3					d_1	d_2	d_3	
1	80	50	3	+1	+1	+1	3,06	0,27	94,29	0,5793	0,4524	0,9383	0,24592
2	20	50	3	-1	+1	+1	2,97	0,03	98,18	0,8000	0,9905	0,9958	0,78910
3	80	10	3	+1	-1	+1	3,08	0,65	91,15	0,4773	0,0002	0,5632	0,00005
4	20	10	3	-1	-1	+1	3,03	0,53	96,48	0,8000	0,1000	0,9863	0,07960
5	80	50	1	+1	+1	-1	3,00	0,03	97,20	1,0000	0,9905	0,9917	0,98231
6	20	50	1	-1	+1	-1	3,00	0,30	96,43	1,0000	0,3676	0,9859	0,36273
7	80	10	1	+1	-1	-1	3,01	0,79	94,42	0,9851	0	0,9435	0
8	20	10	1	-1	-1	-1	3,08	0,90	92,63	0,4773	0	0,8157	0

$$S_{D_{об.}} = \sqrt{\frac{\sum_{n=1}^{N}(Y_{n_i} - \bar{Y}_i)^2}{N-1}}. \qquad (11)$$

$$S_{D_{об.}} = \sqrt{\frac{0{,}0078313}{4-1}} = 0{,}0511;$$

$$V = \frac{0{,}0511 \cdot 100}{0{,}6921} = 7{,}38\,\%.$$

The meaning of the variation coefficient of the eligibility function Dgen., being 7,38%, shows the satisfactory quality of an experimental unit [17].

Full factorial experiment (FFE) using the DCC Box-Wilson plan of 2k type. To plan the experiment optimization the receiving $Co_3(PO_4)_2 \cdot 8H_2O$ a matrix was made which included the variation of three managing factors: x_1 – the synthesis temperature, °C; x_2 – H_3PO_4 concentration, % (wt.); x_3 – the synthesis duration, hours.

Judging the data received when estimating the synthesis results reproducibility, there were not found additional disturbing factors were not revealed and the given data are full enough to regulate the process of synthesis. Table 4 shows the plan of FFE.

Table 5 shows the FFE results and calculation data of partial and united Harrington's desirability function.

The analysis of the results showed that experiment №5 was characterized by practical ideal meaning of Harrington's desirability function that is 0,98231. It means that the next search is not needed because even without building the polynomial model the aim was achieved which was to get a AR reagent having the following conditions of the synthesis such as temperature is 80°C; H_3PO_4 concentration – 50% (wt.); the synthesis duration is 1 hour.

Another confirmation of the quality of a received product fully meeting the demands of reagent purity is the results of researches using the X-ray method. They showed that the X-ray graphics of the composition received in experiment 5 fully meets the demands of standard [18].

So using the FFE method and following Box-Wilson plan $Co_3(PO_4)_2 \cdot 8H_2O$ syntheses were done taking into consideration three response functions in order to use of the received results to build a polynomial model. The response functions different in physical content and size were united using Harrington's desirability function that at the same time considered the requirements to a chemical content of a synthesis as well as to the output of a ready product. It was showed that at the stage of performing FFE we managed to receive the product that 98,23% met the possible meaning of Harrington function of Harrington. That is why there was no need to build a polynomial model and its extreme point. The period of search and financial expenses of conducting the experiment is greatly reduces while doing it with the synthesis conditions of $Co_3(PO_4)_2 \cdot 8H_2O$.

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АННОТАЦИЯ

Войтенко Л.В., Копилевич В.А. *Оптимизация условий синтеза $Co_3(PO_4)_2 \cdot 8H_2O$ реактивной чистоты с использованием функции желательности Харрингтона//Биоресурсы и природопользование. – 2014. – **6**, № 3–4. – С. 27–35.*

С целью ускорения и оптимизации условий синтеза $Co_3(PO_4)_2 \cdot 8H_2O$ реактивной квалификации выполнено математическое планирование эксперимента по плану Бокса-Уилсона. В качестве критерия оптимизации трех функций отзыва использовали функцию желательности Харрингтона, которая позволила объединить разные по физическому содержанию требования к объекту исследования и технологии его получения. Уже при выполнении плана реплики, даже без поиска оптимума и составления полиномиальной модели, были определены условия, полностью удовлетворяющие поставленным требованиям. Степень соответствия состава полученного продукта требованиям к реактиву квалификации "химически чистый" доказана методами химического анализа и РФА.

АНОТАЦІЯ

Л.В. Войтенко, В.А. Копілевич. *Оптимізація умов синтезу $Co_3(PO_4)_2 \cdot 8H_2O$ реактивної чистоти з використанням функції бажаності Харрінгтона //Біоресурси і природокористування. – 2014. – **6**, № 3–4. – С. 27–35.*

З метою прискорення і оптимізації умов синтезу $Co_3(PO_4)_2 \cdot 8H_2O$ реактивної чистоти виконано математичне планування експерименту за планом Бокса-Уілсона. В якості критерію оптимізації трьох функцій відгуку використано функцію бажаності Харрінгтона, яка об'єднала різні за фізичним змістом вимоги до хімічного складу реактиву та технології його одержання. Вже при виконанні плану репліки навіть без проведення пошуку оптимуму та складання поліноміальної моделі було встановлено умови, що повністю задовольнили вимогам. Ступінь відповідності складу продукту вимогам до реактиву кваліфікації "хімічно чистий" доведено методами хімічного аналізу та РФА.