

Investigation of the Process of Purification of Wet-Process Phosphoric Acid and Production of Concentrated Phosphoric Fertilizers Based on it

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Authors' contributions

This work was carried out in collaboration among all authors. Author GQ designed the study, performed the statistical analysis and wrote the protocol; author IS wrote the first draft of the manuscript; author BS translated and corrected this manuscript. Author RN managed the analyses of the study; author BM managed the literature searches. All authors read and approved the final manuscript.

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ABSTRACT

Fluoride compounds have the most harmful effects on the environment. The main source of fluoride in the soil is phosphorus-containing fertilizers. Purification of wet-process phosphoric acid (WPPA) from fluorine will significantly reduce its content in phosphorous-containing fertilizers and improve the quality of the resulting products. The aim of the study is to reduce the content of fluorine and sulfates in the wet-process phosphoric acid by introducing calcium carbonate, dolomite or phosphorite into the finished extraction pulp before filtration and obtaining high-quality calcium and magnesium-containing phosphate fertilizers based on it. Phosphates were determined by differential photometric, fluorine – ionometric, calcium, magnesium, aluminum, and iron-complexometric, and sulfates-by weight methods. For the first time, scientifically-based data were obtained on the simultaneous reduction of the content of fluorine and sulfates in WPPA from phosphorites of Central

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Kyzylkum and the production of calcium-magnesium phosphate fertilizers based on it. Optimal technological parameters of the process of simultaneous defluorination and desulfatation of WPPA from Central Kyzylkum phosphorites with calcium carbonate, dolomite and washed burnt phosphoconcentrate (WCPC) were found by introducing them into the second section of the extractor, into the finished extraction pulp in the amount of 100% for sulfate binding and 100-150% for fluorine binding, as well as obtaining high-quality products based on purified WPPA. The degree of transition of fluorine to the gas phase and phosphogypsum at a rate of 100-150% of calcium oxide for fluorine binding is 86.2-89.4% and its content in the wet-process phosphoric acid decreases from 1.18% to 0.22-0.29%. At the same time, the SO₃ content in the acid decreases from 1.21% to 0.24-0.26%. The filtration rate of the sulfate-phosphate pulp varies slightly and is 807.6-812.6 kg/m²·h by dry residue.

Keywords: Wet-process phosphoric acid; extraction pulp; calcium carbonate; washed burnt phosphate concentrate; defluorination; desulfatation; filtration; calcium and magnesium phosphates.

1. INTRODUCTION

Fluoride compounds have the most harmful effects on the environment. Studies show that fluoride has a negative effect not only on plants, but also on people, animals, and fish, causing various serious diseases [1,2].

Some plants can accumulate significant amounts of fluoride. Thus, tea contains from 57 to 1370 mg of fluoride per 1 kg, and cotton contains up to 4500 mg of fluoride per 1 kg [2]. Moreover, fluoride accumulates in cotton seeds, and in the case of cottonseed oil production, it passes into it. Studies show that when fluoride enters the soil, including with mineral fertilizers, the content of fluoride in the crop increases [3]. The amount of fluorine absorbed by plants increases even more in the presence of nitrogen-phosphorus-potash fertilizers [4].

The main source of fluoride in the soil is phosphorus-containing fertilizers. Thus, apatites and phosphorites contain on average 3.0 and 2.7% fluorine, respectively. Produced on the basis of phosphorites of Central Kyzylkums, wet-process phosphoric acid (WPPA) contains up to 1.5% fluorine. Methods of precipitation from acid with alkali metals in the form of silicofluorides are ineffective, since the acid practically does not contain acid-soluble silicon [5].

During the processing of natural phosphates, the fluorine contained in them is redistributed between the gas, liquid (H₃PO₄) and solid (phosphogypsum) phases. When phosphoric acid is obtained using the dihydrate scheme, 80-85% of the fluorine contained in the raw material (apatite and phosphorite) passes into the acid and remains in fertilizers during its further processing.

Due to the ever-increasing consumption of fertilizers in agriculture, a significant increase in the production of phosphorous fertilizers, especially complex ones, the danger of possible "fluoridation" of soils, plants and water basins becomes obvious [6]. The harmful effect of fluorine compounds entering the atmosphere and ground water on the animal and plant world has been studied quite fully [3,4,7].

Sulfates, chlorides, phosphates, carbonates, and hydroxides of sodium and potassium are used to precipitate fluorine from wet-process phosphoric acid in the form of poorly soluble compounds of alkali metal silicofluorides [8]. These methods are based on the chemical interaction of the above salts with hydrofluoric acid and its soluble salts present in the WPPA. The degree of purification from fluoride reaches up to 90%. However, the degree of purification from fluorine of WPPA from phosphorites of Central Kyzylkums by this method does not exceed 38-40%.

The processes of purification of WPPA from phosphorites of Central Kyzylkum with alkali metal salts are described in detail in the literature. In [9], WPPA was purified sequentially from sulfates and only then from fluorine. The authors investigated in detail the processes of defluorination WPPA on the basis of phosphorite of Central Kyzylkum sulfate, dihydrogen phosphate, the sodium metasilicate and the possibility of increasing the degree of defluorination from 38-40% to 80-85% by the addition of acid silicon dioxide and silicon metasilicate and the technology of defluorination WPPA [10-12].

There are materials on desulfatation of defluorination WPPA with unenriched phosphate raw materials of Central Kyzylkum [13], carbonate, calcium oxide, washed burnt

phosphorite [14-16]. Patented a method of purifying wet-process phosphoric acid from fluoride and sulphate with obtaining forage precipitate [17]. However, materials for simultaneous purification from fluorine and sulfates of sulfuric-phosphoric acid extraction pulp in the preparation of WPPA of Central Kyzylkum were not found.

Currently, the demand for phosphate raw materials in the world is 190 million tons or 43 million tons per year for P_2O_5 . According to forecasts, the demand for phosphate raw materials is expected to grow to 1.3 million tons by 2020 and to 2 million tons by 2030. By 2050, the demand for phosphate raw materials will grow to 220 million tons or up to 70 million tons of P_2O_5 [18].

Due to the growing demand of agriculture for phosphorous fertilizers and the decrease in stocks of high-quality phosphate raw materials year after year, attracting low-grade phosphate raw materials for processing into phosphorous fertilizers is currently one of the urgent tasks.

Neutralization of wet-process phosphoric acid with natural carbonate raw materials allows to increase the number of products by 4-5% than when obtaining ammophos.

Produced phosphorous fertilizers contain a large amount of fluorine (up to 4%) and this to a certain extent has a negative impact on the agrochemical efficiency of fertilizers. In this regard, studies were conducted on the purification of WPPA from fluoride.

Purification of WPPA from fluorine is carried out in three ways [8,17]: Desfluorination by acid evaporation; conversion of fluorine compounds in acid into insoluble compounds and their isolation from WPPA; extraction of fluoride compounds from acid by organic reagents. Purification of wet-process phosphoric acid by these methods requires additional reagents, volumetric equipment and large capital expenditures.

Therefore, research aimed at finding methods of cleaning wet-process phosphoric acid from fluoride and sulphate are relevant.

2. MATERIALS AND METHODS

Calcium carbonate – limestone, composition (mass %): CaO = 54.09, MgO = 1.07, R_2O_3 = 0.19, SO_3 = 0.09, CO_2 = 43.65, H_2O = 0.29, insoluble residue = 0.62, dolomite composition (mass %): CaO = 32.36; MgO = 18.68; CO_2 =

45.76; R_2O_3 = 0.53; H_2O = 1.0, insoluble residue = 1.34, washed and burned phosphate concentrate (WBPC) of Central Kyzylkum, composition in mass %: P_2O_5 = 26.20; CO_2 = 3.08; CaO = 57.64; MgO = 0.60; R_2O_3 = 0.79; SO_3 = 2.22; F = 2.88; insoluble residue = 1.54 and WPPA, composition in mass %: P_2O_5 = 17.23 and 20.15; CaO = 0.32 and 0.41; MgO = 0.66 0 0.82; Fe_2O_3 = 0.36 and 0.28; Al_2O_3 = 0.81 and 0.33; F = 1.18 and 1.32; SO_3 = 1.21 and 2.22.

Precipitation of fluorine and sulfates from the extraction pulp was carried out with calcium carbonate (chalk) dolomite and WBPC at a rate of 60-150% calcium oxide for the formation of calcium fluoride and 80-100% for the binding of SO_3 .

The experiments were carried out on a laboratory, continuous model installation consisting of a two-section stainless steel extractor with an isolated electric heating layer, equipped with electric mixers and dispensers of acids and phosphate flour. The capacity of the plant is 150 g of phosphate raw materials, the working volume of the extractor is 2.5 liters.

The decomposition of phosphorite was carried out with a mixture of sulfuric and phosphoric acids circulating in the dehydrate mode. The system worked without a circulation of the pulp. Before the operation both sections of the extractor was filled with solvent extraction of the pulp obtained in terms of the dehydrate mode from standard raw materials. The mixing speed in the first reactor was 120-140 rpm, and in the second 80-100 rpm, the process duration was 3 hours. The ratio of the liquid and solid phases is 2.5, the excess of the sulfate ion in the liquid phase in terms of SO_3 was 3.3-3.5 g/100 ml. Filtration was performed on a Buchner funnel through a filter cloth at a discharge of 400 mm Hg.

The residence time of the extraction pulp in the first section of the reactor is 1 hour, and in the second 2 hours, since the volume of the second reactor is 3 times greater than the first, and the speed of rotation of the agitator is 1.5 times less than in the first reactor. This provides favorable conditions for the process of crystal formation.

Chemical analysis of raw materials, intermediate and final products was performed using well-known methods [19-22].

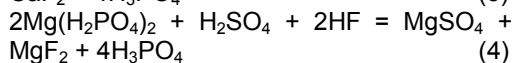
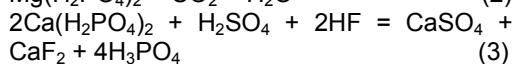
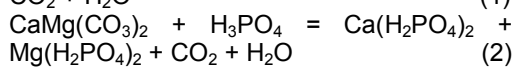
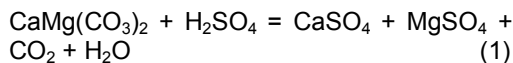
The results are shown in Tables 1,2,3,4 and Figs 1,2.

Table 1. Influence of the norm of calcium carbonate on the chemical composition of WPPA and the phase separation of fluorine

Name of indicators	The total rate of calcium carbonate on the binding of fluorine, % of stoichiometry						
	-	60	80	100	120	140	150
Norm of calcium carbonate for binding of free H ₂ SO ₄ , % of stoichiometry	-	80	100	100	100	100	100
Chemical composition of the wet-process phosphoric acid, mass. %							
P ₂ O ₅	17.23	17.28	17.25	17.19	17.10	16.98	16.90
CaO	0.32	0.39	0.54	0.88	1.40	2.21	2.73
MgO	0.66	0.72	0.71	0.68	0.68	0.67	0.67
SO ₃	1.21	0.47	0.28	0.26	0.25	0.25	0.24
R ₂ O ₃	1.17	1.09	1.08	1.08	1.07	1.08	1.07
F	1.18	0.62	0.46	0.29	0.24	0.23	0.22
suspensions	0.29	0.31	0.27	0.24	0.21	0.18	0.15
The degree of transition of fluorine, %							
In phosphogypsum	41.3	69.1	74.5	81.8	84.9	85.2	85.4
In gas phase	5.1	5.1	4.9	4.4	4.2	4.1	4.0
Total	46.4	74.2	79.4	86.2	89.1	89.3	89.4

3. RESULTS AND DISCUSSION

Table 1 shows that when obtaining WPPA without the introduction of calcium carbonate, dolomite and WCPC, the degree of transition of fluorine to the gas phase is 5.1% of the total amount in phosphorite, 41.3% passes into phosphogypsum and 53.6% of fluorine remains in the WPPA. When calcium carbonate or dolomite is introduced into the extraction pulp, the following reactions may occur:



Magnesium fluoride, in contrast to calcium fluoride, is more soluble in acids, and reacts with strong acids, in particular, with sulfuric acid, to form magnesium sulfate and hydrogen fluoride by reaction:



This process is complex, since monophosphate and magnesium sulfate, which are highly soluble in WPPA, can interact with calcium carbonate to form calcium sulfate and monophosphate. However, magnesium carbonate will immediately react with phosphoric acid to form magnesium monophosphate. This is confirmed by the fact

that the magnesium content in the WPPA is almost unchanged and remains at the level of 0.66-0.72% Table 1. With the introduction of calcium carbonate and the formation of calcium fluoride, the amount of fluorine released into the gas phase decreases from 5.1 to 4.0%, which indicates the release of its main amount at the beginning of the process in the first reactor. The total degree of transition of fluorine to the gas phase and phosphogypsum during decomposition with the introduction of calcium carbonate when normal of 100-150% is 86.2-89.4%. The content of fluorine in the WPPA is 0.22-0.29%, which is 4.1-5.4 times less than in the case without the introduction of calcium carbonate.

The degree of defluorination of WPPA at a rate of 100-120% CaO in the form of CaCO₃ for the fluorine content is 75.4-80.5%, and desulfation of 78.5-79.3% at a rate of 100% Figs. 1 and 2. The figures show that when the rate of calcium carbonate exceeds 120% based on the available content of fluorine and sulfates in the acid, the degrees of defluorination and desulfation change slightly. The amount of fluorine remaining in the WPPA is 10.6-13.8% of the total amount in phosphorite. When normal of calcium carbonate changes to the available fluorine in phosphorite from 60 to 100%, an additional transition of fluorine to phosphogypsum is observed by 27.8-40.5%. Increasing the rate of calcium carbonate to 120-150% increases the degree of transition of fluorine to the solid phase by only 3.1-3.6%. The excess content of calcium carbonate during the

deposition of fluorine from the WPPA is spent on the formation of calcium sulfate, with an excess of sulfuric acid, and monocalcium phosphate, by interaction with phosphoric acid. Table 1 shows that the SO_3 content decreases from 1.21% to 0.24-0.26%, and the calcium oxide content increases from 0.32% to 2.73%. In this case, the coefficients of decomposition, extraction, washing and yield are 98,8-99,1%, 96,1-96,3%, 99,0-99,2% and 95.3%, respectively, for the norms of calcium carbonate for fluorine binding from 60 to 150% and excess sulfuric acid from 80 to 100% Table 2.

The filtration rate of phosphogypsum is relatively high and is 807.6-812.6 $\text{kg/m}^2\cdot\text{h}$ in dry matter.

Table 3 and Figs. 1 and 2 show the results of defluorination and desulfatation of WPPA with washed burnt phosconcentrate from Central Kyzylkums. In contrast to the use of calcium carbonate in the acid purification process, when using WBPC, the P_2O_5 content in the production WPPA increases from 19.85-20.15% to 20.15-21.89%, the CaO content in the acid is 1.46-2.15% with a norm of 120-150% for the formation of calcium sulfate, The MgO and SO_3 contents are approximately the same, the fluorine content decreases in the acid to 0.31%. Figs. 1 and 2 show comparative data on the degree of defluorination and desulfatation with calcium carbonate, dolomite, and WBPC. The figures show that with the introduction of calcium carbonate, the degree of defluorination is higher by 2.4-2.7%, and the degree of desulfatation is worse by 1.7-2.3% with a norm of 100-120% CaO for defluorination and desulfatation.

When fluorine and sulfates are deposited with dolomite raw materials at 100% CaO norm relative to fluorine and 100% CaO norm relative to sulfates, the SO_3 content in the WPPA

decreases by 0.39%, and F by 0.35%. When the stoichiometric ratio of dolomite to fluorine increases to 120%, the SO_3 content in the product decreases to 0.26%, and F is 0.24%.

With an increase in the norm of dolomite raw materials, there is no further decrease in the content of fluorine and sulfates.

After filtration of the extraction pulp, a purified solution of WPPA is formed, the composition (mass %): $\text{P}_2\text{O}_5 = 17.38$; CaO = 1.09; MgO = 1.07; $\text{SO}_3 = 0.26$.

After filtration of the extraction pulp using limestone, a purified WPPA of the composition (mass %) is formed: $\text{P}_2\text{O}_5 = 17.38$; CaO = 1.19; MgO = 0.77; $\text{SO}_3 = 0.26$; F = 0.23.

The conducted studies have shown the principal possibility of simultaneous defluorination and desulfatation of WPPA from phosphorites of Central Kyzylkums by introducing calcium carbonate and WBPC into the extraction pulp. The optimal rate of calcium carbonate and WBPC is 100-120% CaO for the formation of calcium fluoride and 100% CaO for the formation of calcium sulfate. At the same time, the content of sulfates decreases from 1.21% to 0.24-0.26%, fluorine from 1.18% to 0.22-0.29%, the degree of transition of fluorine to the gas phase during extraction decreases from 5.1% to 4.2-4.4%, and in phosphogypsum increases from 41.3% to 81.8-84.9%.

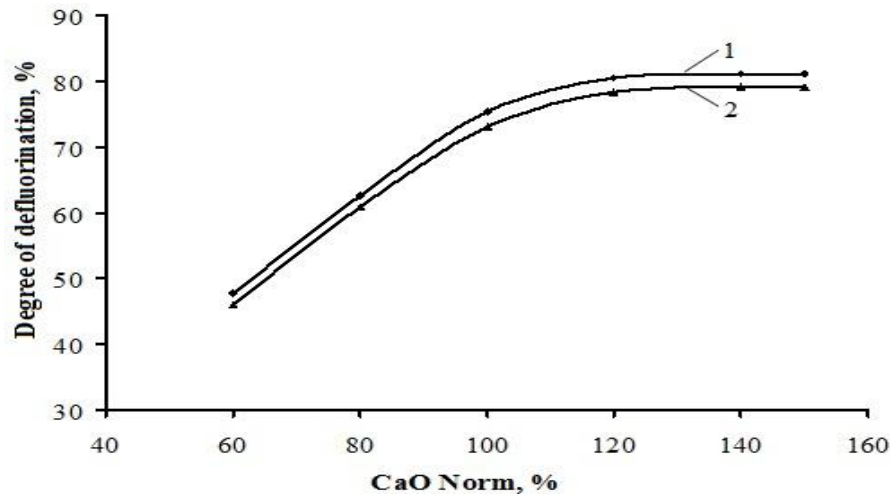
A two-stage process of neutralization of WPPA with natural calcium and calcium-magnesium carbonates (limestone, dolomite) was studied in order to obtain high-quality de-fluorinated phosphorous fertilizers containing calcium and magnesium phosphates.

Table 2. Technological indicators of production of defluorination and desulfurized wet-process phosphoric acid

Name of indicators	The total rate of calcium carbonate on the binding of fluoride and sulphate, the stoichiometry						
	-	140	180	200	220	240	250
$K_{\text{decomp.}}$, %	99.1	99.0	98.9	98.9	98.9	98.8	98.8
$K_{\text{extractions}}$, %	96.3	96.2	96.2	96.1	96.1	96.1	96.1
K_{washings} , %	99.0	99.1	99.1	99.2	99.2	99.2	99.2
$K_{\text{the output}}$, %	95.3	95.3	95.3	95.3	95.3	95.3	95.3
Pulp density (ρ), g/cm^3 at 25°C	1.24	1.24	1.24	1.24	1.25	1.25	1.25
Pulp viscosity (η), centipoise (cst) at 25°C	3.10	3.42	3.52	3.61	3.68	3.72	3.73
Filtration rate of the extraction pulp, $\text{kg/m}^2\cdot\text{h}$	815.4	812.6	811.7	810.6	809.5	808.3	807.6

Table 3. Influence of the WCPC norm on the chemical composition of WPPA and the phase distribution of fluorine

Name of indicators	The rate of phosphate on the binding of fluorine, % of stoichiometry						
	-	60	80	100	120	140	150
Norm of washed burnt phosconcentrate for binding of free H ₂ SO ₄ , % of stoichiometry	-	80	100	100	100	100	100
Chemical composition of WPPA, mass. %							
P ₂ O ₅	20.15	20.87	21.28	21.13	21.71	21.81	21.89
CaO	0.41	1.04	1.17	1.26	1.55	1.94	2.15
MgO	0.82	0.85	0.86	0.85	0.88	0.89	0.88
SO ₃	2.22	0.90	0.61	0.47	0.48	0.48	0.48
R ₂ O ₃	0.61	0.63	0.64	0.63	0.65	0.66	0.66
F	1.32	0.74	0.56	0.36	0.30	0.31	0.31
suspensions	0.25	0.34	0.31	0.25	0.27	0.28	0.19
The degree of transition of fluorine, %							
In phosphogypsum	41.3	69.1	74.5	81.8	84.9	85.2	85.4
In gas phase	5.1	5.1	4.9	4.4	4.2	4.1	4.0
Total	46.4	74.2	79.4	86.2	89.1	89.3	89.4

**Fig. 1. Influence of the norm of calcium carbonate (1) and WBPC (2) on the degree of desfluorination**

The first stage was carried out in the process of obtaining purified wet-process phosphoric acid, by introducing calcium carbonate or dolomite in the amount of 100% CaO for fluorine deposition and 120% CaO for SO₃ deposition.

The second stage of neutralization was performed with a stoichiometric norm of CaCO₃ (pH = 2.8-3.5), corresponding to the neutralization of purified WPPA to calcium monophosphate and magnesium monophosphate. The resulting pulp was

evaporated to a content of 30-35% H₂O and dried at a temperature of 100-105°C.

Technological indicators of the process of neutralization of WPPA with limestone and dolomite raw materials, chemical compositions of the obtained intermediate and final products are shown in Table 4.

When neutralizing WPPA purified from fluorine and sulfates with limestone or dolomite at the stoichiometric normal, a phosphate pulp is formed with a SO₃ content of 0.24% and a content of 0.22 and 0.23% F, respectively Table 4.

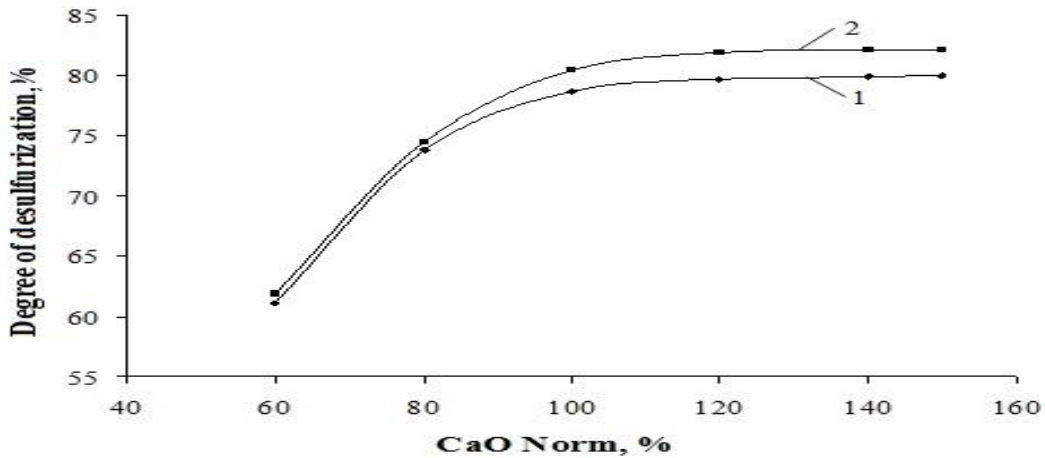


Fig. 2. Influence of the norm of calcium carbonate (1) and WBPC (2) on the degree of desulfatation

Table 4. The technological parameters and chemical composition of the products of neutralization of wet-process phosphoric acid with limestone and dolomite raw materials

No	Indicators	Chemical composition, %			
		Superphosphate pulp		Finished product-calcium and magnesium phosphates	
1.	Stoichiometric rate of CaCO ₃ for CaO at the 1st and 2nd stages of the process, %:				
	In the 1st stage of F	100	120	100	120
	In the 1st stage of SO ₃	100	100	100	100
	In the 2nd stage-to neutralize WPPA	100	100	100	100
Neutralization by dolomite					
2.	P ₂ O _{5t}	16.47	16.77	50.08	51.99
3.	P ₂ O _{5 acc.c.a.}	15.72	16.40	47.75	50.83
4.	P ₂ O _{5 w.s.}	14.78	15.32	44.86	47.43
5.	CaO _t	4.36	4.18	13.25	12.96
6.	MgO _t	2.84	2.85	8.64	8.84
7.	SO _{3t}	0.36	0.24	1.02	0.74
8.	F _t	0.32	0.23	0.98	0.70
9.	H ₂ O	68.04	68.71	2.81	2.95
10.	(P ₂ O _{5ac.c.a.} :P ₂ O _{5t})x100 %	95.45	97.79	95.35	97.77
11.	(P ₂ O _{5w.s.} :P ₂ O _{5t})x100%	89.74	91.35	89.58	91.22
Neutralization of limestone					
12.	P ₂ O _{5t}	16.86	17.07	49.56	49.84
13.	P ₂ O _{5 acc.c.a.}	16.30	16.78	47.90	48.97
14.	P ₂ O _{5 w.s.}	15.38	15.90	45.11	46.29
15.	CaO _t	6.84	6.80	20.10	19.85
16.	MgO _t	0.33	0.37	0.98	1.07
17.	SO _{3t}	0.27	0.24	0.82	0.70
18.	F _t	0.26	0.22	0.75	0.63
19.	H ₂ O	67.08	66.95	3.26	3.51
20.	(P ₂ O _{5ac.c.a.} :P ₂ O _{5t})x100%	96.68	98.30	96.65	98.25
21.	(P ₂ O _{5w.s.} :P ₂ O _{5t})x100%	91.22	93.15	91.02	92.88

As a result of evaporation and drying of the suspension, fertilizers of the composition (wt. %): P_2O_5 (total-t.) = 51.99 and 49.84; P_2O_5 (acceptable by citric acid-ac.c.a.) = 50.83 and 48.97; P_2O_5 (water soluble) = 47.43 and 46.29; CaO = 12.96 and 19.85; MgO = 8.84 and 1.07; SO_3 = 0.74 and 0.70; F = 0.70 and 0.63; H_2O = 2.95 and 3.51.

In this case, the ratio $[P_2O_5(ac.c.a.):P_2O_5(t.)] \times 100$ is 97.77 and 98.25%, and the ratio $[P_2O_5$ (water soluble-w.s.): $P_2O_5(t.)$] X 100 is 91,22 and 92 and 88%.

The resulting fertilizers are part of high-grade, environmentally effective fertilizers such as double superphosphate.

When neutralizing WPPA purified from fluorine and sulfates with limestone or dolomite at the stoichiometric normal, a phosphate pulp is formed with a SO_3 content of 0.24% and a

content of 0.22 and 0.23% F, respectively Table 4.

As a result of evaporation and drying of the suspension, fertilizers of the composition (wt. %): P_2O_5 (t.) = 51.99 and 49.84; P_2O_5 (ac.c.a.) = 50.83 and 48.97; P_2O_5 (w.s.) = 47.43 and 46.29; CaO = 12.96 and 19.85; MgO = 8.84 and 1.07; SO_3 = 0.74 and 0.70; F = 0.70 and 0.63; H_2O = 2.95 and 3.51.

In this case, the ratio $[P_2O_5(ac.c.a.):P_2O_5(t.)] \times 100$ is 97.77 and 98.25%, and the ratio $[P_2O_5(w.s.):P_2O_5(t.)] \times 100$ is 91,22 and 92 and 88%.

The resulting fertilizers are high-quality, environmentally effective, such as fertilizers such as double superphosphate.

The block diagram of material flows of calcium and magnesium phosphate fertilizers showed in Fig. 3.

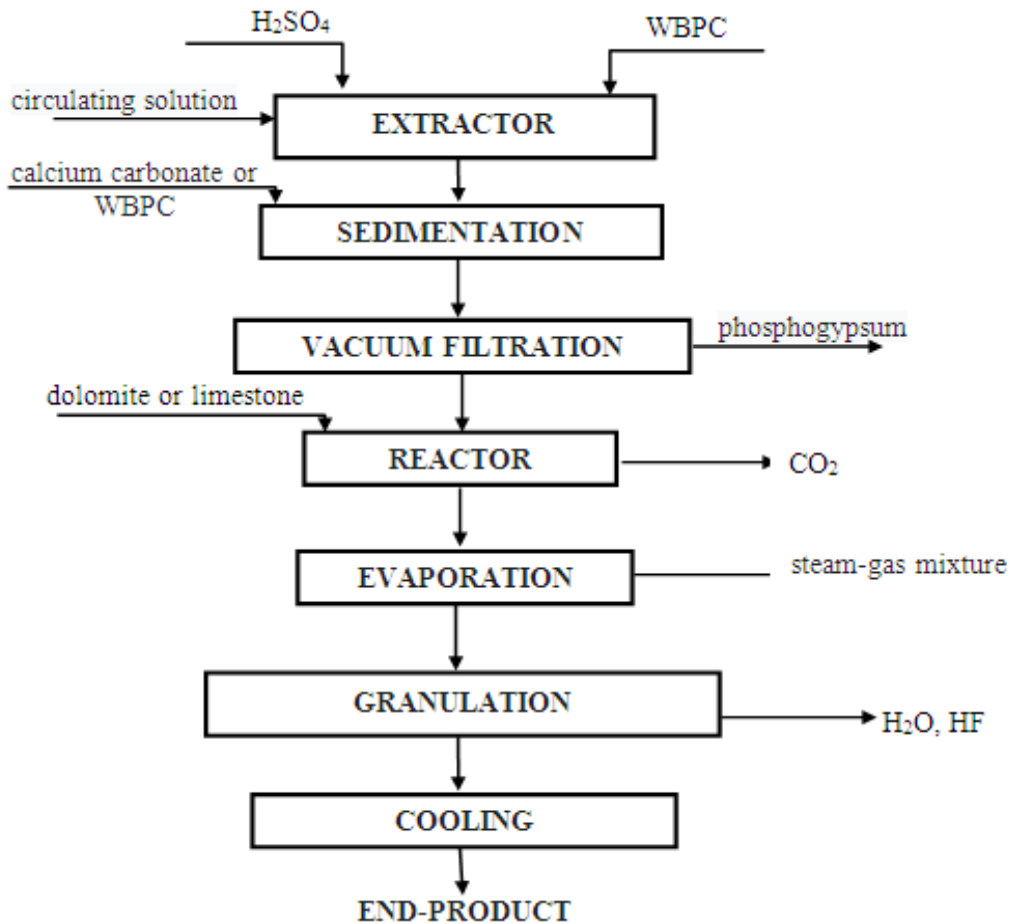


Fig. 3. Block diagram of material flows of calcium and magnesium phosphate fertilizers

4. CONCLUSION

Thus, the conducted studies have shown the possibility of simultaneous defluorination and desulfatation of wet-process phosphoric acid from phosphorites of Central Kyzylkum by introducing a calcium-containing reagent into the finished sulfuric-phosphoric acid pulp in the amount of 100% of the reagent for sulfates and 120% based on fluorine. Neutralization of purified WPPA with dolomite at the rate of 70-100% for the formation of monocalcium and monomagnesium phosphates allows obtaining fluorinated phosphorous fertilizers with a composition close to double superphosphate.

COMPETING INTERESTS

Authors have declared that no competing interests exist.

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