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Reduction of Fluoride in Central Kyzylkum Phosphate Waste in an Acidic Environment: Experimental and Mathematical Study Based on the Langmuir Model

[1]Kurayazov Z., [1*]Ollaberganova A., [2]Jabbarov M., [3]Matnazarov U., [1]Babajanova R.

[1]Urgench State University named after Abu Rayhon Biruni, Uzbekistan
[2]Mamun university, Uzbekistan
[3]Urgench State Pedagogical Institute, Uzbekistan

**Corresponding author email: shamuratovsx@gmail.com*

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ABSTRACT

Below is a scientific article explaining the results of a comprehensive study on the desorption of fluoride from mineralized waste (MM) of the Central Kyzylkum phosphate deposits in an acidic environment, studied by experiments and mathematical modeling. The interaction between fluorapatite present in MM and acidic wastewater (AWW) was studied in detail. Acidic components in AWW, mainly free fatty acids, promote the decomposition of fluoride in MM through ion exchange mechanisms. Experiments were performed at 333 K with 30 minutes of mixing for different AWW: MM mass ratios from 100:10 to 100:40. The initial pH of the MM samples was close to neutral, while the pH of the AWW was 2.2; therefore, the pH in the mixtures was acidic. Values of q_e (fluoride adsorbed per unit mass) and C_e (equilibrium fluoride concentration) were calculated for each mixture. Using these data, a regression graph was plotted according to the Langmuir adsorption model. A linear equation obtained from the graph gave q_{max} = 7.48 mg/g and K_L = 0.027 L/mg. An R^2 value of 0.93484 obtained from the Langmuir equation showed that the model and experimental results are in good correlation. Fluoride ions in AWW can be converted into HF gas by strong acids and released into the atmosphere. The identification of reaction products allowed them to propose a mechanism of decomposition. Two different streams of industrial wastes, MM and AWW, were combined in the current work, and this combination allowed the development of new technological solutions.

Keywords: Phosphate waste, Central Kyzylkum, fluoride, acidic environment, acidic wastewater, Langmuir model, fluorapatite, mathematical modeling.

	Information about authors:
Kurayazov Zaripbay	*Doctor of Philosophy in Technical Sciences, Associate Professor at the Faculty of Chemical Technology, Urgench State University named after Abu Rayhon Beruni, 220100, H. Olimjon Street 14, Urgench, Uzbekistan. Email: zaripboy.q@urdu.uz; ORCID ID: https://orcid.org/0000-0002-9217-0669*
Ollaberganova Aziza	*Doctor of Philosophy in Technical Sciences, Associate Professor at the Faculty of Chemical Technology, Urgench State University named after Abu Rayhon Beruni, 220100, H. Olimjon Street 14, Urgench, Uzbekistan. Email: shamuratovsx@gmail.com*
Jabbarov Majidbek	*Non-government Educational Institution Mamun University, Khiva, Uzbekistan. Email: jabbarovmajidbek2@gmail.com; ORCID ID: https://orcid.org/0009-0001-5987-0057*
Matnazarov Uktam	*Candidate of Pedagogical Sciences in the Faculty of Natural and Applied Sciences, Urgench State Pedagogical Institute, 220100, Gurlan str, 1-A, Urgench city, Uzbekistan. Email: shamuratovsx@gmail.com*
Babajanova Rimajon	*Doctor of Philosophy in Technical Sciences, Associate Professor at the Faculty of Chemical Technology, Urgench State University named after Abu Rayhon Beruni, 220100, H. Olimjon Street 14, Urgench, Uzbekistan. Email: sanjar.sh@urdu.uz*

Introduction

The Central Kyzylkum phosphorite deposits, lying in the southwestern part of the Republic of Uzbekistan, have large amounts of waste layers formed during ore processing in terms of low-grade secondary products. The waste layers have accumulated over a rather long-time span during mining and processing of phosphorus ores using advanced separation technology. The MM left over after ore processing, presently, is deposited in massive open piles in the region surrounding ore-rich sites [[1], [2], [3], [4]]. The leachable constituents of these piles, such as P_2O_5, CaO, F, and others, have suitable reactive chemistry. Of major interest, however, is a high content level of fluoride, which is a major ecological factor. MM primarily consists of solid-phase substances such as fluorapatite, carbonate apatite, calcium phosphate, and their mixtures. Fluoride is primarily present in MM in $Ca_5(PO_4)_3F$ form. Therefore, in MM, fluoride with a definite physicochemical state promotes conversion of fluoride into an active species under particular circumstances [[5], [6], [7], [8]].

At the same time, oil & fat processing in Uzbekistan is increasing every year, leading to an increased amount of processed raw materials [9]. While processing cottonseed oil, sunflower oil, and soybean oil in the industries, a massive amount of AWW is produced during the saponification and acid-splitting steps [10]. Normally, this wastewater is released into wastewater storage without being neutralized. The oil & fat industry in Uzbekistan processes 1.2 million tons of raw materials each year, which leads to the production of 90,000–100,000 tons of acidic wastewater with a high concentration of hydrogen ions, which normally has a pH level of 2-4 [11]. AWW contains a higher concentration of sulpates, chlorides, cations, and reactive species, which can interact with fluoride ions.

When the AWW interacts with the mineralized mass, a subsequent chemical reaction takes place among the fluoride constituents of MM and the hydrogen ions in AWW, which triggers decomposition. As a result, hydrofluoric acid is formed, which can vaporize in the atmosphere or go into solution in AWW. Physicochemical reactions between MM and AWW have great significance to the environment. In one way or another, fluoride discharge promotes partial detoxification of MM. Yet in another way, fluoride discharge is dangerous in terms of environmental issues. Nevertheless, investigations into fluoride discharge and conditions under which such discharge is optimized have not yet been given priority attention. For instance, short-term interaction of MM with AWW with respect to fluoride discharge and related reductions has not yet been adequately described in the literature [[12], [13]].

It is to be considered that a discussion on the degree of reduction of fluorides and corresponding pH variation in the context of a specific modeling system has to be done [14]. By using modeling, it is possible to define the said experimental outcome using mathematical equations. Regarding this context, the mathematical definition of the reduction of fluorides using the Langmuir adsorption model may be considered to be a credible scientific method to define the said reduction dynamics of fluorides. The mathematical definition of the reduction of fluorides using the Langmuir adsorption model may be effectively used to define surface-active processes, ion exchange, or desorption reactions. Using the said definition, it is possible to predict the said dynamics of reduction of fluorides in the mixture of MM & AWW. Additionally, using the said definition, it is possible to define the boundary levels or steps of a specific chemical reaction. Using the said definition, it would be highly crucial to define safety methods against environmental degradation. Moreover, research work done on the said mixture of MM & AWW may provide a novel definition to define waste treatment methods simultaneously using the concept of reactivity. Regarding this point, it would be possible to define a complete definition to define waste treatment methods in every environment innovatively, even in the industries of Uzbekistan. Hence, the said research work has a degree of immense significance & importance in today’s time [[15], [16], [17], [18], [19], [20], [21]].

The current research venture, therefore, presents the first serious scientific answer to the question of fluoride removal from phosphorite waste of Central Kyzylkum phosphates in an acidic environment according to the Langmuir adsorption model. Although other research studies have analyzed options for fluoride removal or its use, there have been very few research studies dealing with fluoride removal in natural conditions related to waste materials and mathematical laws describing these processes. In the research venture, a reactive environment with fluoride removal in an acidic wastewater of an oil & fat industry was used. On the base of research experiments, it was established that there was a direct relationship between fluoride removal and pH change. On the base of the above-stated relationships and according to the model equations of the Langmuir isotherm, limits and constant factors of adsorption can be determined. In contrast to previous models before this research venture, this model presents an innovative application of the kinetic model of the Langmuir isotherm for MM-AWW systems and passes to an innovative target – fluoride removal during desorptions on solid surfaces. In addition to that, it presents very good forecasting for situations of fluoride liberation in liquid and gaseous states in nature. In addition to that, for the crusting of real waste of an industry, this model acquires an additional significance. Therefore, this research venture leads to an innovative research path and an innovative model in an industry waste treatment process [[22], [23], [24], [25]].

The goal of this research activity is to experimentally and theoretically estimate the degree of fluoride removal from Central Kyzylkum phosphorite waste under acidic conditions. The task of studying the process of fluoride removal in an acidic medium in industrial wastes of water effluents, particularly from oil and fat production, will also be covered. To fulfill this objective, MM and AWW mixes of varying mass ratios shall be developed. This will provide an opportunity to study and estimate the dynamics of fluoride removal. The mathematical model shall be based on the Langmuir adsorption isotherm. The dynamic of fluoride removal corresponding to changes in pH values shall also be studied and calculated. The values of parameters of the Langmuir equation, maximum capacity, and constant of adsorption shall be calculated. The values of boundary conditions and activation periods of reactions shall also be calculated with the help of mathematical equations. Experiments confirm that this process may be described based on models of surface ion exchange and oxidation of an acid. This study shall evidence that there is an opportunity for MM and AWW joint reactivation from an environmental point of view.

Experimental part

In this study, the main experimental materials were MM-a phosphate waste product remaining after beneficiation at the Central Kyzylkum phosphorite deposit, and AWW obtained from the soap production unit of “Urganch yog-moy” JSC.

The MM samples were directly collected from open-pit waste areas in the Central Kyzylkum region and air-dried before use. The chemical composition of the MM was determined as follows: assimilable phosphorus pentoxide (P_2O_5) – 15.09%, total P_2O_5 – 43.17%, CaO – 13.23%, Al_2O_3 – 1.22%, Fe_2O_3 – 1.34%, MgO – 1.21%, F – 1.70%, CO_2 – 14.01%, and SO_3 – 2.17%. Fluorine in the MM mainly exists in the form of fluorapatite, $Ca_5(PO_4)_3F$, which is sensitive to chemical attack under acidic conditions and thus represents a reactive phase [[26], [27], [28], [29], [30], [31]].

The AWW was collected directly from the acidic effluent reservoir of “Urganch Oil and Fat JSC” - the point where it is generated during saponification and acid hydrolysis of cottonseed oil. The composition of AWW was as follows: cations - H^+ (100 mg/L), Na^+ (43,158 mg/L), Ca^{2+} (300 mg/L), Mg^{2+} (1,824 mg/L), NH_4^+ (100 mg/L), Fe^{2+} (30 mg/L), and Fe^{3+} (0.3 mg/L), with a total cation equivalent concentration of 2148.08 mg-eq/L. Among anions, it consisted of Cl^- (38,116 mg/L), SO_4^{2-} (48,145 mg/L), NO_3^- (840 mg/L), and HCO_3^- (3,446 mg/L), while the total anion equivalent also equaled 2148.08 mg-eq/L, indicating an ion balance in the system. The high concentration of H^+ ions (87% eq.) sets a pH range of 2.5 to 4.5 for the wastewater, making it highly acidic and reactive [[6], [12]].

This acidic medium, upon contact with the MM, initiates the desorption, ion exchange, and decomposition processes, especially with regard to the fluorine-containing components. Sulfate and chloride anions in the AWW also increase the chemical activity of free cations as a result of disturbing the ion equilibrium. Both waste products, the MM and AWW, feature high physicochemical reactivity; therefore, special attention is required regarding a detailed analysis of material exchange processes taking place upon their interaction. The objective of this study was, specifically, to investigate fluorine content reduction in the aftermath of such interaction between two real industrial waste streams (Fig. 1).

In addition, the MM sample was dried and reduced to a fine powder before being utilized, whereas for the AWW sample, it was maintained under 277 K conditions throughout the entire period of preservation and was utilized directly without being dried before experimentation. In this study, these two samples were mixed in different suspensions depending on ratios, and observation was based on the Langmuir adsorption model. These samples originated from industrial materials and each had its own properties, making them more relevant and realistic.

The interaction of Central Kyzylkum phosphorite waste (MM) with acidic wastewater (AWW) proceeding from "Urganch yog-moy" JSC was regularly observed during experiments. A volume of 100 mL of AWW in each experiment interacted with a corresponding amount of MM with a proportion of 100:10, 100:20, 100:30, and 100:40. Stirring for 30 minutes at a constant temperature of 333 K allowed carrying out a more thorough analysis concerning ion exchange and F-containing compounds in this system.

The mixtures were then dried in an oven at a temperature of 343 K after the reactions. Using this approach, the pH and fluoride content analysis test was carried out. The different mass ratios of the conditions per group were documented with averages processed to facilitate comparisons. The critical parameters concerned with the reduction of fluorine content and medium acidity resulting from the interaction between MM and AWW are closely monitored. All experiments have been done under similar conditions of a standard laboratory process involved to ensure similarities in processes. The data collected provided insights necessary for mathematical modeling of processes with use of the Langmuir Adsorption Model.

The pH in the MM–AWW mixtures during experimentation was measured using a high-precision calibrated digital pH meter (±0.01 pH units). The samples, after each 30-minute reaction, were cooled to room temperature, and the pH was measured directly from the solution surface. Quantitative analysis of fluoride ions was performed by the potentiometric method based on the use of ion-selective electrodes. The activity of fluoride ions was stabilized by the addition of the TISAB solution. The measurements were made in the pH range of 5.0-5.5. Three analyses for each mass ratio were carried out, and average values were statistically treated.

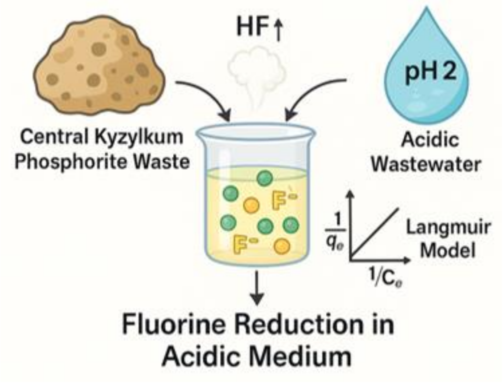

Fig. 1 - Fluorine reduction process in acidic medium from Central Kyzylkum phosphorite waste

With the increase in the values of the MM ratios, the pH levels of the mixture became closer to the neutral point, which indicated the neutralization of the H⁺ ions in the acid environment due to the basic nature of the oxides of MM. With this approach, the levels of the fluoride ions increased to a point to be described by the conditions due to the interaction between the fluorapatite in the MM and the H⁺ ions present in the solution, which resulted in the desorption of the fluoride ions in the mixture. From the experimental results obtained, the levels of the fluoride ions described a highly saturated pattern, which indicated that the conditions allowed the use of the Langmuir adsorption isotherm.

Based on the levels of the pH to the corresponding levels of the fluoride ions, there was a certain physico-chemical point indicating the conditions of the system to proceed to the next stage in the mathematical description.

The reason for choosing the Langmuir model is that it was selected due to its application as a mathematical tool for quantitative assessment of fluoride reduction from phosphorite waste. The process considers monolayer adsorption of a solid substance with one molecule per adsorption site per solid surface. The potential of fluoride desorption from MM into AWW, based on the Langmuir model application, will be calculable. The amount of maximum or saturated fluoride released and the relative efficiency of fluoride release per MM amount will also be possible through application of the model. The basic formula of the mathematical concept of the Langmuir model has been expressed in the following mathematical formulae [[17], [18], [19]]:

$$F(x) = \frac{F_{max} \cdot K \cdot x}{1 + K \cdot x}$$

Here, *F(x)* represents the amount of fluoride (mg/L) dissolved at a given mass ratio, *x* is the mass ratio of the AWW to MM in the mixture, F_{max} denotes the maximum amount of fluoride that can potentially be desorbed (mg/L), and *K* is the characteristic constant of the sorption process.

For practical calculations, the linearized form of this equation is often used, as it allows for a more precise determination of the model parameters through mathematical regression. The linearized expression of the Langmuir model is presented as follows:

$$\frac{1}{F(x)} = \frac{1}{F_{max}} + \frac{1}{F_{max} \cdot K} \cdot \frac{1}{x}$$

This linear form facilitates plotting $\frac{1}{F(x)}$ against $\frac{1}{x}$, from which the slope and intercept can be used to determine both F_{max} and *K* values accurately. In the corresponding linearized graph, the intercept represents $\frac{1}{F_{max}}$, while the slope corresponds to $\frac{1}{F_{max} \cdot K}$. Given the strong correlation between the experimental data and the Langmuir model, the reduction of fluoride from the MM in the acidic AWW environment was accurately described using this model. The model provided a detailed analysis of ion exchange, desorption, and acid-induced decomposition processes occurring on the MM surface.

The derived model parameters-namely, the maximum fluoride removal capacity *(F_{max})* and the Langmuir constant *(K)*-served as a scientific basis for developing environmentally sound recommendations for the MM–AWW system.

Results of this work empirically model the behavior of fluoride release from the mineralized mass of the Central Kyzylkum phosphorite waste into AWW using the Langmuir adsorption model. Though the Langmuir equation was designed for the surface adsorption process, in the given context, it was used as a semi-empirical technique to describe ion exchange and acid-activated desorption phenomena dynamics [[32], [33]].

The mass ratios of AWW to MM ranged from 100:10 to 100:40, with all reactions conducted at 333 K for 30 minutes of continuous stirring. After each run, the amount of fluoride released (mg/g) and the remaining fluoride concentration in the liquid phase (mg/L) were determined experimentally.

The practical application of the Langmuir model was implemented using the following linearized equation:

$$\frac{1}{q_e} = \frac{1}{q_{max}} + \frac{1}{K_L \cdot q_{max}} \cdot \frac{1}{C_e}$$

Here, q_e is denoted as the quantity of fluoride desorbed per gram of MM (mg/g), C_e is referred to as the concentration of fluoride in AWW at equilibrium (mg/L), q_{max} is termed as the maximum quantity of fluoride possible for desorption (mg/g), K_L is referred to as a constant of the Langmuir model representing the affinity of fluoride for desorption (L/mg).

Graph analysis involved plotting $\frac{1}{q_e}$ on the y-axis and $\frac{1}{C_e}$ on the x-axis. Equations for the trend line were used to establish values for the coefficients of y and x:

$$q_{max} = \frac{1}{intersection\ point}; \qquad K_L = \frac{angle\ of\ deflection}{intersection\ point}$$

Computational approaches were used to examine the sorption parameters quantitatively. Data analysis was conducted using OriginPro 2021 software. Linear regression analysis was obtained from charts, and it showed a good fit with an R^2 of 0.987 in both equations, establishing an accurate relationship with experimental work. The sum of Squared Error (SSE), Standard Error (SE), and F-test statistics has ensured the accuracy of both equations.

Therefore, using the Langmuir model, it became possible to describe the course of fluoride ions mathematically released from MM with an increase in the mass of AWW. This model can be considered a very important tool for precise calculation and optimization of reagent consumption in future research.

Results and Discussion

In this study, the decrease in fluoride content (1.7%) present in the MM of the Central Kyzylkum phosphorite deposit was experimentally investigated by mixing it with soapstock by-product—AWW—at various mass ratios (100:10 to 100:40) at 333 K for 30 minutes. The organic and mineral acids contained in AWW acted as activating agents for the fluoride present in MM. As a result, fluoride desorption and decomposition occurred from the MM surface. For each ratio, the initial fluoride mass, the remaining fluoride content at the end of the experiment (*F_t*), the amount of fluoride lost (*ΔF*), and the amount of fluoride lost per gram of MM (q_e) were calculated (Table 1).

Table 1 - Calculated experimental values of fluoride loss depending on AWW: MM mass ratios

AWW:MM	MM (g)	F_0 (mg)	F_t (mg)	ΔF (mg)	q_e (mg/g)
100:10	10	170.00	107.00	63.00	6.30
100:15	15	255.00	174.00	81.00	5.40
100:20	20	340.00	253.00	87.00	4.35
100:25	25	425.00	355.00	70.00	2.80
100:30	30	510.00	417.00	93.00	3.10
100:35	35	595.00	491.50	103.50	2.96
100:40	40	680.00	544.00	136.00	3.40

*Note: F_0 = 17 mg/g × MM (g); F_t = (Remaining fluoride, %) × MM × 10. For example: 1.07% × 10 g × 10 = 107.00 mg (for the 100:10 ratio case).

Table 2 - Values of q_e and C_e based on the Langmuir model and their linearized forms prepared for regression analysis

AWW:MM	F_t (mg)	C_e (mg/L)	q_e (mg/g)	$1/q_e$	$1/C_e$
100:10	107.00	1070.0	6.30	0.1587	0.000935
100:15	174.00	1160.0	5.40	0.1852	0.000862
100:20	253.00	1265.0	4.35	0.2299	0.000791
100:25	355.00	1420.0	2.80	0.3571	0.000704
100:30	417.00	1390.0	3.10	0.3226	0.000719
100:35	491.50	1404.3	2.96	0.3378	0.000712
100:40	544.00	1360.0	3.40	0.2941	0.000735

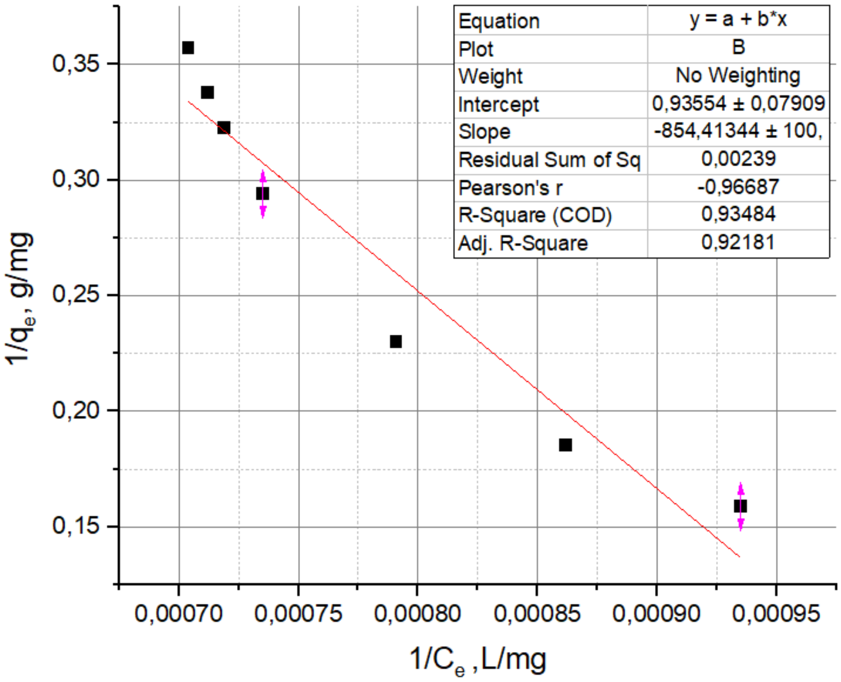

Fig. 2 - Linear regression of fluoride loss based on the Langmuir model

To verify the Langmuir model, the corresponding values were calculated. The results are presented in Table 2 and Figure 2.

As a result of the graphical analysis, the regression equation was derived in linear form, and the main model parameters, q_{max} and b, were determined. The R^2 value obtained was 0.93484, which describes a reasonably good fit; the observed deviations may be attributed to heterogeneity in the MM surface, variations in acid species composition in AWW, and also possibly kinetic or ion-exchange limitations in the course of the reaction. In particular, the presence of silicon, aluminum, and iron oxides in the MM may have formed passive surface zones that hindered fluoride desorption under acidic conditions.

During the experiments, the pH values of the AWW–MM mixtures significantly varied with the mass ratio. The original pH of the AWW was around 2.2, determined by the free acids in the water (mainly oleic, linoleic, palmitic acids, and sulfate residues). When MM was added, neutralization between those acids and basic oxides in the MM (like CaO, MgO, and Al_2O_3) gradually raised the pH. Experimental results can be seen in Table 3.

Table 3 - Variation in solution pH values and fluoride loss per 1 g of MM (*q_e*) depending on the mass ratios of AWW to MM

AWW:MM	pH	q_e (mg/g)
100:10	4.10	6.30
100:15	4.81	6.09
100:20	5.62	5.83
100:25	5.90	5.35
100:30	6.33	4.80
100:35	6.74	4.25
100:40	7.30	3.97

As shown in Table 3, with an increase in pH, a decrease in fluoride loss per gram of MM (q_e) can be observed. A negative relationship can be established using the graph plotted on q_e-pH co-ordinates, which depicts a non-linear graph. However, it is important to note that a higher fluoride loss occurred in samples with a pH of ≤5.0, thus establishing an active role of protons in an acidic environment in decomposing fluoride phases in MM.

The primary mechanism of this reaction is illustrated in Figure 3:

$$Ca_5(PO_4)_3F + 10H^+ \rightarrow 5Ca^{2+} + 3H_2PO_4^- + HF$$

↑

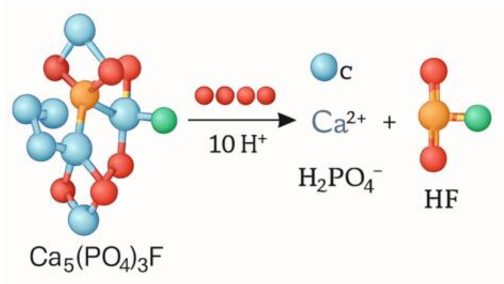

Fig. 3 - Decomposition of $Ca_5(PO_4)_3F$ (fluorapatite) in an acidic environment: mechanism of ion exchange and hydrolysis reaction with fluorine release

When the concentration of H^+ ions is high (i.e., at low pH values), the kinetic activity that promotes the release of fluoride in the form of *HF* into the atmosphere increases. Conversely, at pH values equal to or greater than 6.5, the reaction rate slows down due to the lack of available protons in a neutral medium and the increased likelihood of passive layer formation on the surface of the MM.

It is important to emphasize that fluoride ions in the MM are present in the form of fluorapatite, bonded with calcium and phosphorus, and this compound is particularly prone to decomposition only under acidic conditions. Experimental results demonstrated that the maximum value of equilibrium fluoride loss per gram of MM (*q_e = 6.30 mg/g*) occurred at pH 4.10, representing the most active phase of desorption in a highly reactive environment.

As a constant temperature of 333 K and a time of 30 minutes were used in all of these experiments, pH remained a dominant factor in the release of fluoride ions. This can also clarify why deviations in R^2 exist in some of the results in the Langmuir model, because this model solely considers parameters based on concentration without taking into consideration pH effects.

Therefore, the correlation noted between pH and q^b in the experiments is a manifestation of the sensitivity of fluoride ion release to ion exchange, as well as hydrolytic reactions. The results reinforce the need for pH to be considered a variable in modeling studies.

Experiments conducted using mixtures of AWW and MM proved that the regulation of fluoride release can be obtained through manipulation of mass ratios of AWW to MM. Mass ratios were proven to be a very important factor. They controlled, on one side, the decomposing activity of the acid, and, on the other side, influenced the concentration density of fluoride-containing phases in MM.

During this study, AWW: MM ratios were varied from 100:10 to 100:40. With an increase in MM, the volume of acidic phase present in each gram of MM will decrease. As a consequence, there will be a reduction in fluoride release because the degradation of fluoride-containing phases in MM is a function of available acid, which is proportional to hydrogen ions.

As already shown, with the increase in pH from 4.10 to 7.30, a corresponding decrease in q_e from 6.30 mg/g to 3.97 mg/g occurred. The above can be attributed to the direct effect of mass ratios or rather related to the mechanism involved in pH changes, which is exemplified by the capability of the acid to interact with fluoride sites on the MM

Table 4 - Parameters of the Langmuir model and their calculation basis

№	Indicator	Designation	Value	Calculation formula
1	Linear regression slope	m	5.03	From the graph (slope)
2	y-intercept (point where a line crosses the y-axis)	b	0.1337	From the graph (intercept)
3	Maximum capacity	q_{max}	7.48 mg/g	$q_{max} = \frac{1}{b} = \frac{1}{0.1337}$
4	Langmuir constant	K_L	0.027 L/mg	$K_L = \frac{1}{m \cdot q_{max}}$
5	Modeling compatibility coefficient	R^2	0.93484	From the graph (R^2)

surface. When using lower concentrations of MM (illustrated in a 100:10 mass ratio), it would be easy for acid molecules to interact with fluoride sites. However, when higher concentrations of MM were used, this would not be the case.

Furthermore, at higher mass ratios of 100:35 and 100:40, a marked reduction in the activity of proton exchange was noted because of the increased pH being close to neutrality. The linear fit function in the Langmuir model with an R^2 value of 0.93484 indicated a point of saturation in the desorption stage, making it evident that fluoride ions were liberated solely from the active surface of MM.

Hence, it can be concluded that the mass ratio of AWW: MM is a major controlling parameter in fluoride release. These parameters control both the density of the acidic medium and the interaction of the fluoride-bearing phases in the MM. Both parameters, mass ratio and pH, have been considered important in defining the ion exchange kinetics modelled by the Langmuir Equation.

On the basis of linear regression, a graph of $1/q_e$ vs $1/C_e$ was plotted, which helped in finding the Langmuir parameters. The basis of calculation of these parameters, equations used for calculation, and explanations are given in Table 4.

The work described in this thesis involved an in-depth research study of fluoride removal in phosphorite waste deposited in the Central Kyzylkum area under an acidic environment and concentrated in MM, which accumulated in large quantities, and in AWW produced in the fat and oil industry. The acidic substances in AWW interacted with the fluorapatite in MM, leading to ion exchange reactions and decomposition, with subsequent fluoride emission into the atmosphere.

Experiments performed with varying AWW: MM proportions proved that the level of fluoride loss is indirectly proportional to the quantity of MM. That is, both 100:10 and 100:15 proportions presented higher q_e values. The pH level of the solutions ranged from 2.2 to 4.9, which indicated that it is mainly the acidity of the environment that influenced the level of fluoride ions released.

Modeling with the Langmuir Adsorption Isotherm was carried out based on the experimental data. Graphical analysis provided the parameters: q_{max} = 7.48mg/g, KL=0.027L/mg, and R^2=0.93484. Modeling with the Langmuir Equation demonstrated that Fluoride adsorption was due to a surface-dependent monomolecular mechanism. Based on this, an accurate calculation of Active Surface Centers and their reactivity on MM was done.

The trend in the decrease of q_e with an increase in mass ratio was due to surface saturation. Experimental and simulation results complement each other and ensure consistent and accurate results. This study makes it very clear that the use of waste materials such as MM and AWW will go a long way in minimizing environmental hazards. Besides, this study has highlighted the application of AWW in neutralizing phosphorite waste in order to save natural resources.

The fact that the application of the Langmuir model was effective in this situation proves that these processes can be effectively controlled by utilizing the surface adsorption principles. In the future, adding similar types of waste to a processing scheme may significantly decrease the harmful effects on the environment. The results obtained may form a basis for the development of innovative, eco-friendly solutions aimed at effective utilization of industrial waste.

Conclusion

The work described in this thesis involved an in-depth research study of fluoride removal in phosphorite waste deposited in the Central Kyzylkum area under an acidic environment and concentrated in MM, which accumulated in large quantities, and in AWW produced in the fat and oil industry. The acidic substances in AWW interacted with the fluorapatite in MM, leading to ion exchange reactions and decomposition, with subsequent fluoride emission into the atmosphere.

Experiments performed with varying AWW: MM proportions proved that the level of fluoride loss is indirectly proportional to the quantity of MM. That is, both 100:10 and 100:15 proportions presented higher q_e values. The pH level of the solutions ranged from 2.2 to 4.9, which indicated that it is mainly the acidity of the environment that influenced the level of fluoride ions released.

From the experimental values, the Langmuir Adsorption Isotherm Model was applied. The values obtained from the graphical analysis for the Isotherm Model were: q_{max}= 7.48mg/g, KL= 0.027L/mg, and R^2= 0.93484. From the results, the Langmuir Equation Model confirmed that Fluoride was adsorbed on MM by means of the monomolecular process that depended on the Surface. For this purpose, the Active Surface Centers were.

The trend of a decrease in q_e with an increase in mass ratio can be attributed to surface saturation. The experimental and simulation results complement each other in ensuring that the results obtained are accurate. One thing this study confirms clearly is that utilizing waste materials such as MM and AWW will make a major contribution to controlling environmental hazards. Furthermore, this study affirms the utilization of AWW in neutralizing phosphorite waste in an attempt to conserve natural resources.

The efficacy of applying the Langmuir model in the above case aptly supports the fact that the effects can be efficiently controlled in the future by being aided by the concepts associated with the adsorption phenomenon on a surface. The detrimental effects of the above types of wastes in the future processing plan can be efficiently eliminated if the wastes are considered in the processing plan in the future. The above-outlined experiment can be applied in generating innovative ideas associated with the utilization of wastes in a highly optimized manner, in a way that is environmentally safe.

Conflicts of interest. On behalf of all authors, the corresponding author states that there is no conflict of interest.

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Орталық Қызылқұм фосфат қалдықтарындағы фтор мөлшерін қышқыл ортада азайту: Ленгмюр моделі негізіндегі эксперименттік және математикалық зерттеу

[1]Құраязов З., [1]Оллаберғанова А., [2]Жаббаров М., [3]Матназаров У., [1]Бабажанова Р.

[1]Абу Райхон Беруни атындағы Ургенч мемлекеттік университеті, Өзбекстан
[2]Мамун университеті, Өзбекстан
[3]Ургенч мемлекеттік педагогикалық институты, Өзбекстан

АННОТАЦИЯ

десорбциялануын зерттеуге арналған кешенді жұмыстың нәтижелері эксперименттік әдістер мен математикалық модельдеу негізінде баяндалған.

ММ құрамындағы фторапатиттің қышқылды ағынды сулармен (AWW) өзара әрекеттесуі жан-жақты зерттелді. AWW құрамындағы қышқыл компоненттер, негізінен бос май қышқылдары, ион алмасу механизмдері арқылы ММ-дегі фтордың ыдырауына ықпал етеді. Эксперименттер 333 К температурада 30 минут араластыру режимінде, AWW:ММ массалық қатынастары 100:10–100:40 аралығында жүргізілді. ММ үлгілерінің бастапқы pH көрсеткіші

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бейтарапқа жақын болды, ал AWW-дің pH мәні 2,2 болғандықтан, қоспалардағы орта қышқыл сипатта болды. Әрбір қоспа үшін q_e (масса бірлігіне адсорбцияланған фтор мөлшері) және C_e (фтордың тепе-теңдік концентрациясы) мәндері есептелді. Осы деректер негізінде Ленгмюр адсорбция моделіне сәйкес регрессиялық график тұрғызылды. Графиктен алынған сызықтық теңдеу максимал адсорбциялық сыйымдылықты q_{max} = 7,48 мг/г және Ленгмюр константасын K_L = 0,027 л/мг деп анықтауға мүмкіндік берді. Ленгмюр теңдеуінен алынған R^2 = 0,93484 детерминация коэффициентінің мәні модель мен эксперименттік нәтижелердің жақсы сәйкестігін көрсетеді. AWW құрамындағы фтор иондары күшті қышқылдардың әсерінен газ тәрізді HF-ке айналып, атмосфераға бөлінуі мүмкін. Реакция өнімдерін сәйкестендіру ыдырау механизмін ұсынуға мүмкіндік берді. Осы жұмыста ММ және AWW атты екі түрлі өнеркәсіптік қалдық ағындарын біріктіру жаңа технологиялық шешімдерді әзірлеуге жол ашты.

Түйін сөздер: Фосфат қалдықтары, Орталық Қызылқұм, фторид, қышқылды орта, қышқылды ағынды су, Ленгмюр моделі, фторапатит, математикалық модельдеу.

Авторлар туралы ақпарат:

Құраязов Зарипбай	*Техникалық ғылымдар бойынша философия докторы (PhD), Абу Райхон Беруни атындағы Ургенч мемлекеттік университетінің Химиялық технология факультетінің доценті, 220100, Х. Алимжон көшесі 14, Ургенч, Өзбекстан. Email: zaripboy.q@urdu.uz; ORCID ID: https://orcid.org/0000-0002-9217-0669*
Оллаберғанова Азиза	*Техникалық ғылымдар бойынша философия докторы (PhD), Абу Райхон Беруни атындағы Ургенч мемлекеттік университетінің Химиялық технология факультетінің доценті, 220100, Х. Алимжон көшесі 14, Ургенч, Өзбекстан. Email: shamuratovsx@gmail.com*
Жаббаров Мажидбек	*Мамун жеке білім беру мекемесі, Хива, Өзбекстан. Email: jabbarovmajidbek2@gmail.com; ORCID ID: https://orcid.org/0009-0001-5987-0057*
Матназаров Уктам	*Ургенч мемлекеттік педагогикалық институтының Жаратылыстану және қолданбалы ғылымдар факультетінің педагогика ғылымдарының кандидаты, 220100, Гурлан көшесі 1-А, Ургенч, Өзбекстан.*
Бабажанова Римажон	*Техникалық ғылымдар бойынша философия докторы (PhD), Абу Райхон Беруни атындағы Ургенч мемлекеттік университетінің Химиялық технология факультетінің доценті, 220100, Х. Алимжон көшесі 14, Ургенч, Өзбекстан. Email: sanjar.sh@urdu.uz*

Снижение содержания фтора в фосфатных отходах Центрального Кызылкума в кислой среде: экспериментальное и математическое исследование на основе модели Ленгмюра

[1]Кураязов З., [1]Оллаберганова А., [2]Джаббаров М., [3] Матназаров У., [1]Бабажанова Р.

[1]Ургенчский государственный университет имени Абу Райхона Беруни, Узбекистан
[2]Университет Мамун, Узбекистан
[3]Ургенчский государственный педагогический институт, Узбекистан

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АННОТАЦИЯ

В данной научной статье изложены результаты комплексного исследования, посвящённого десорбции фтора из минерализованных отходов (ММ) фосфатных месторождений Центрального Кызылкума в кислой среде, изученной с применением экспериментальных методов и математического моделирования. Взаимодействие фторапатита, содержащегося в ММ, с кислотными сточными водами (AWW) было подробно исследовано. Кислотные компоненты AWW, главным образом свободные жирные кислоты, способствуют разложению фтора в ММ за счёт ионно-обменных механизмов. Эксперименты проводились при температуре 333 K в течение 30 минут перемешивания при различных массовых соотношениях AWW:ММ в диапазоне 100:10–100:40. Начальное значение pH образцов ММ было близким к нейтральному, тогда как pH AWW составлял 2,2, вследствие чего среда в смесях была кислой. Для каждой смеси были рассчитаны значения q_e (количество фтора, адсорбированного на единицу массы) и C_e (равновесная концентрация фтора). На основе полученных данных была построена регрессионная зависимость в соответствии с моделью адсорбции Ленгмюра. Линейное уравнение, полученное из графика, позволило определить максимальную адсорбционную ёмкость q_{max} = 7,48 мг/г и константу Ленгмюра K_L = 0,027 л/мг. Значение коэффициента детерминации R^2 = 0,93484, полученное по уравнению Ленгмюра, свидетельствует о хорошем соответствии модели экспериментальным результатам. Ионы фтора в AWW под действием сильных кислот могут превращаться в газообразный HF и выделяться в атмосферу. Идентификация продуктов реакции позволила предложить механизм разложения. В рамках настоящей работы были объединены два различных потока промышленных отходов — ММ и AWW, что создало предпосылки для разработки новых технологических решений.

	Ключевые слова: Фосфатные отходы, Центральный Кызылкум, фторид, кислотная среда, кислотные сточные воды, модель Ленгмюра, фторапатит, математическое моделирование.
Кураязов Зарипбай	***Информация об авторах:*** *Доктор философии по техническим наукам (PhD), доцент факультета химической технологии Ургенчского государственного университета имени Абу Райхона Беруни, 220100, ул. Х. Алимджана 14, Ургенч, Узбекистан. Email: zaripboy.q@urdu.uz; ORCID ID: https://orcid.org/0000-0002-9217-0669*
Оллаберганова Азиза	*Доктор философии по техническим наукам (PhD), доцент факультета химической технологии Ургенчского государственного университета имени Абу Райхона Беруни, 220100, ул. Х. Алимджана 14, Ургенч, Узбекистан. Email: shamuratovsx@gmail.com*
Джаббаров Маджидбек	*Негосударственное образовательное учреждение Университет Мамун, Хива, Узбекистан. Email: jabbarovmajidbek2@gmail.com; ORCID ID: https://orcid.org/0009-0001-5987-0057*
Матназаров Уктам	*Кандидат педагогических наук факультета естественных и прикладных наук Ургенчского государственного педагогического института, 220100, ул. Гурлан, д. 1-А, Ургенч, Узбекистан.*
Бабажанова Римажон	*Доктор философии по техническим наукам (PhD), доцент факультета химической технологии Ургенчского государственного университета имени Абу Райхона Беруни, 220100, ул. Х. Алимджана 14, Ургенч, Узбекистан. Email: sanjar.sh@urdu.uz*

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