Обгрунтовано доцільність застосування реагентних і фізико-хімічних методів для очищення висококонцентрованих стоків підприємств м'ясопереробної промисловості. Показано, що ефективність реагентного очищення стічних вод із застосуванням дешевих природних реагентів, зокрема кальцію оксиду, збільшується під впливом акустичних коливань ультразвукового діапазону

Ключові слова: висококонцентровані стічні води, механічне очищення, флотація, коагулянти, кавітація, біологічне очищення

Обоснована целесообразность использования реагентних и физико-химических методов для очистки высококонцентрированных стоков предприятий мясоперерабатывающей промышленности. Показано, что эффективность реагентной очистки сточных вод с использованием дешевых природных реагентов, и кальция оксида в отдельности, увеличивается под влиянием ультразвуковых акустических колебаний

Ключевые слова: высококонцентрированные сточные воды, механическая очистка, флотация, коагулянты, кавитация, биологическая очистка

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RESEARCH INTO PROCESSES OF WASTEWATER TREATMENT AT PLANTS OF MEAT PROCESSING INDUSTRY BY FLOTATION AND COAGULATION

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1. Introduction

Environmental protection is one of the priority tasks of today. The water medium ranks first by the intensity of anthropogenic impact on the environment. Despite the fact that about 71 % of the Earth's surface is covered by waters of the world ocean, the need for high quality drinking water increases every year. Less than 4 % of the total volume of water is fresh, suitable for domestic needs and drinking. However, this water is constantly polluted by insufficiently purified or completely untreated waters, which after getting into natural water bodies alter their composition, decrease self-purification ability of natural ecosystems. The indicated wastewaters cause an increase in the content of organic substances, nitrogen, phosphorus and heavy metals compounds. These compounds degrade water quality and cause oxygen deficiency in water bodies. This has a negative impact on natural ecosystems, leading to their degradation or even destruction.

Given the above, development of effective technology of purification of wastewater with high content of biogenic elements will reduce the technogenic burden on the environment, in particular on natural water bodies.

2. Literature review and problem statement

Among industrial enterprises, which heavily pollute the environment, the impact of food industry companies is significant. First of all, it concerns enterprises that process agricultural raw materials, especially those of cattle-breeding. These enterprises are characterized not only by high water consumption (they occupy one of the first places among other industries), but also by significant dumping of used and contaminated wastewater into the environment: water, soil, and atmosphere. Some of these enterprises, in the best case, are equipped with primitive ineffective local treatment facilities, but most plants do not have even them. Wastewater contains residues of feed, kitchen salt, detergents and disinfectants, nitrogen and phosphorus compounds, alkalis, acids, and pathogenic microflora. Composition and volume of wastewater vary very widely. It depends on the type and composition of raw materials, produce range, seasonal fluctuations in production power, which are typical for most food industry enterprises, in particular meat processing. In addition, qualitative and quantitative indicators of wastewater are influenced by the composition of detergents and disinfectants, used for providing sanitary conditions and washing technological equipment. The quality of wastewater deteriorates significantly in the absence of procedures of disposal waste of processing animal raw materials (internal organs, blood, lymph, feathers, etc.).

These wastewaters contain large amount of organic substances (amount of chemical consumption of oxygen varies within 1500...25000 mgO_2/dm^3), suspension (particularly fats), and pH index may vary within a wide range (4.5...12.5). The mode of formation and discharge of these wastewaters is characterized by considerable irregularity (from 5 to 150 m^3 per day). The average composition of wastewaters of enterprises of food industry is given in Table 1 [1].

Data in Table 1 show that enterprises of food industry are characterized by high values of such indicators as suspended substances (SS), chemical consumption of oxygen (CCO), and biological consumption of oxygen (BCO_5). The highest values of these indicators are inherent in wastewaters of meat processing enterprises [2]. In addition to the high values of CCO and BCO_5, these wastewaters are characterized by a very high content of fats and water-soluble salts of fatty acids. Thus, mechanical methods are used at the initial stage of purification. They include settling [3], centrifuging, filtering [4], etc. Gravitational purification by settling requires significant amounts of production equipment due to the very low sedimentation rate of dispersed particles. Filter treatment of wastewater at meat processing enterprises is often ineffective due to the buildup of insoluble fats on filter surfaces, which dramatically increases their hydraulic resistance. Physical-chemical methods, such as coagulation, [5] or electric coagulation [6], are used for deeper removal of highly dispersed particles. However, in case of a significant content of highly dispersed lipid particles coagulation does not provide proper wastewater treatment. Lipid particles are also able to block electrodes by deposits of fat, which causes overexertion of electrode processes. The choice of the method of mechanical treatment depends on physical and chemical properties of suspension, regularity of wastewaters arrival and their volumetric consumption. Usually, mechanical and physical-chemical treatment is followed by biological (aerobic and non-oxide) treatment [7]. However, in most wastewaters, the ratio between biogenic elements (BCO_5:N:P) is unfavourable for their treatment by biological methods. To provide standard indicators in processes of biological treatment, it is necessary to apply the equipment with considerable working volumes that requires considerable production areas and the electricity costs.

For each specific case, one of the enumerated methods is chosen, or they are combined [8]. However, even a combination of several specified above methods often does not provide achievement of standard indicators in terms of the content of contaminants.

The above determines the search of new ways of effective solution of the problem of treatment of highly concentrated wastewater.

3. The aim and tasks of research

The aim of present work is to develop the technology of treatment of highly concentrated wastewater of meat processing enterprises.

To achieve the set goal, the following tasks were to be solved:

– to explore the efficiency of using flotation for removal of pollutants;

– to study the influence of acoustic oscillations of ultrasound (US) range on the effectiveness of chemical reagents, in particular calcium hydroxide, in the process of wastewater treatment;

– to select the main stages of treatment of highly concentrated wastewater.

4. Materials and methods of examining the technology of treatment of highly concentrated wastewater of meat processing enterprises

The study was carried out using actual industrial wastewaters (Table 2), which were preliminary purified from roughly dispersed particles on a coarse sand filter.

Flotation purification from highly dispersed particles was carried out in the pillared flotation plant of the volume of 0.7 dm^3 with a separator. For reagent treatment, we used the reactor with capacitive type of volume of 1.0 dm^3 with a stirrer with the adjustable number of turnovers.

The influence of acoustic oscillations of the US-range on the reactive activity of reagents, including suspension of $Ca(OH)_2$, and the process of wastewater treatment were examined using the US-radiator of magnetostrictive type "Ultrasonic Disintegrator" UD-20 (Poland). The power of ultrasound radiation is $1...3 \cdot 10^2$ W/m^3, frequency is 22 kHz.

Table 1

Average composition of wastewaters of some enterprises of food industry

Indicators of wastewater pollution	Enterprises of food industry					
	meat packing plant	poultry factory	milk factory	production of apple juice	breweries	production of starch and syrup
pH	6.2–7.6	6.8–7.0	3.7–9.8	4.5–10.0	5.0–10.5	4.7–7.4
SS*, mg/dm^3	1793	6235	493	1464	1392	831
CCO**, mg O_2/dm^3	3430	6687	4116	7413	4187	941
BCO_5***, mgO_2/dm^3	1788	4682	3547	5145	2596****	650
Ammonium nitrogen mg/dm^3	178	77	7.2	14		6
Phosphates, mg/dm^3	128	175	210	2	19	absent
Lipids, mg/dm^3	483	1341	66	absent	absent	absent

*Note: * – suspended substances; ** – chemical consumption of oxygen; *** – biological consumption of oxygen;**** – BCO_5, value of BCO_n in [1] is missing*

Table 2

Composition of wastewaters* of a meat processing enterprise

Sample number	Quality indicators, mg /dm³							
	pH	SS	CCO	BCO	Fats	Nitrogen	Phosphorus	Coloring
Wastewaters, generated within a week								
1	10,25	1120	1450	810	480	120	125	Light-yellow
2	6,00	940	1070	685	450	95	110	
3	10,20	1210	1380	790	475	115	136	
Wastewaters, generated during washing smoking equipment								
1	12,35	1200	4400	850	720	140	145	Dark-brown
2	13,1	1450	6510	920	800	170	157	
3	12,75	1475	6950	1030	810	180	165	

*Note: * – composition of wastewaters after a fat trap*

Calcium hydroxide (air-slaked lime by DSTU B.V.2.7-46-96) in the form of 10...20 % suspension and aqueous solution of sodium hypochlorite (TU U 6-05761620.014-99) were used as reagents. Iron (II) sulfate (TU 2141-002-58318296-2005) in the form of aqueous solution with 20 % by weight was used as a coagulant.

The process of wastewater treatment with the use of suspension of calcium hydroxide was studied by two methods. The first included preliminary activation of CaO suspension in the range of ultrasound radiation. The second one included direct acoustic treatment of wastewater after adding the reagent to it. All studies were performed under isothermal conditions. During the experiment, we observed visually the change in color and transparency of the liquid phase, controlled the change in pH, reduction-oxidation potential (ROP), and determined the magnitude of CCO.

The magnitude of the CCO in the studied media was defined by DSTU ISO 060:2003.

Analysis of the cation-anion composition of wastewater was carried out in accordance with "Procedures of performance of measurements", PPM 081-12-0006-01, PPM 081/12-0004-01, "Guiding standardizing document" GSD 211.1.4.026-95 and DSTU ISO 6332:2003.

Definition of fat content in wastewater was carried out by the Soxhlet method.

Regression analysis of the obtained experimental dependences was performed with the use of standard programs. Based on it, the magnitudes of approximation reliability (R^2) were determined.

5. Results of examining the technology of wastewater treatment at meat processing enterprises

Since it is impossible to reproduce the composition of natural wastewaters, the research was conducted with actual wastewaters, collected at meat processing plant (PP "Miasko", village Domazhyr, Lviv region), which operates using delivered raw materials. Because their composition depends, as it was stated above, on a number of factors, the study was carried out using wastewaters, neutralized by composition (Table 2). As it may be seen from data in Table 2, these wastewaters are characterized by high values of BOC, suspended substances, fats, compounds of biogenic elements, which are difficult to remove and which have a negative impact on the environment.

Considering a high content of suspended highly dispersed substances and fats, we investigated the possibility of using flotation and filtration for their removal.

Flotation of light-yellow and dark-brown wastewaters was carried out separately without correcting their composition, in particular, the pH of the medium. Results of the process of light-yellow wastewater treatment are shown in Fig. 1.

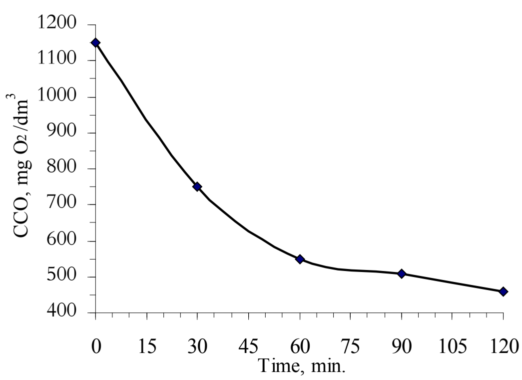

Fig. 1. Dependence of magnitude of chemical consumption of oxygen (CCO) of wastewater of light-yellow coloring on the time during flotation, CCO_0=1150 mg O_2/dm³; pH_0=6.0

During flotation of dark-brown wastewaters within 10...20 min, 50...70 % of the liquid phase were removed from the flotation plant. Clearing the liquid that remained in the flotation plant was not visually observed.

Therefore, based on the previous studies, we concluded that flotation of both wastewaters without preliminary introduction reagents is irrelevant.

Application of iron (II) compounds is caused by the fact that they may simultaneously play the role of a catalyst for oxidation of organic compounds by the oxygen of the air (potential of the system Fe^{3+}/Fe^{2+} is 0.771 V) and of a coagulant.

The results of research into influence of iron (II) sulfate (1 cm³ of 20 % solution per 150 cm³ of wastewater) on the ROP of light-yellow wastewater are shown in Table 3.

The study of the oxidation of dark-brown wastewaters by aeration at existence of iron (II) sulfate was performed with wastewaters at the initial value of the CCO of 4400 mg O_2/dm³ and pH of 12.35. As a coagulant, we used the 20 % solution of iron (II) sulfate. Results of research are shown in Fig. 2, 3.

Table 3

Change in magnitude of pH and reduction-oxidation potential over time in the presence of iron (II) sulfate, pH_0=9,4, ROP_0=0 mV in light-yellow wastewater

Parameters of system	Duration of stirring, min				
	0	10	20	30	40
pH	9,4	7,4	6,9	6,6	6,5
ROP, mV	0	5	18	40	35

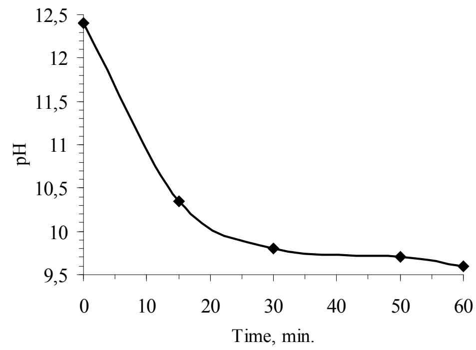

Fig. 2. Dependence of pH magnitude of wastewater of dark-brown coloring on the duration of flotation: $V_{wwaters}$=250 cm^3; V_{coag}=25 cm^3

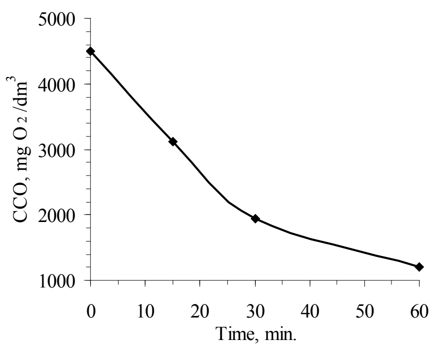

Fig. 3. Dependence of the magnitude of chemical consumption of oxygen (CCO) of wastewater on the aeration duration in the presence of iron (II) sulfate: CCO_0=4400 mg O_2/dm^3; pH_0=12.35 (after correction and addition of iron (II) sulfate, pH=4.5)

Both types of wastewater include soluble compounds of fat row, in particular salts of higher fatty acids. These compounds are formed due to the use of detergents, which contain a variety of surface active substances and alkaline reagents (solution of mixture of sodium and potassium hydroxides). Resulting sodium and potassium salts of fatty acids, that is soap, are the well soluble compounds. Therefore, it is logical to predict that their removal from the dissolved state may be achieved due to formation of insoluble salts, such as calcium salts. These compounds should form sediment, which could be relatively easy to separate from wastewater. Therefore, we studied the effect of calcium compounds, in particular calcium hydroxide, which is a cheap reagent of natural origin, on formation of sediment and the magnitude of BCO. Since calcium hydroxide is characterized by very low solubility (1.5 g/dm^3), to intensify the process with its participation, we applied acoustic oscillations of the US-range [9]. Therefore, further studies were aimed at defining efficiency of application of ultrasonic radiation in the processes of wastewater treatment with calcium hydroxide. The results are shown in Table 4, 5.

Table 4

Influence of ultrasonic acoustic radiation on magnitude of chemical consumption of oxygen (CCO) of wastewater during their treatment with solid CaO (CCO_0=460 mg O_2/dm^3)

Content of CaO in wastewater, %	Magnitude of CCO, mg O_2/m^3/Degree of purification, %	
	Stirring without sonication	Sonication of wastewater with CaO
2.5	420/9	405/12
5.0	360/22	320/30
7.5	315/31	240/48
10.0	280/39	170/63

Table 5

Influence of ultrasonic acoustic radiation on magnitude of chemical consumption of oxygen (CCO) of wastewater during their treatment with suspension of calcium oxide (CCO_0=460 mg O_2/dm^3, duration of process is 20 min)

Content of CaO in wastewater, %	Magnitude of CCO, mg O_2/m^3/Degree of purification, %		
	Stirring without sonication	Sonication of wastewater with reagent	Preliminary sonication of suspension of CaO
10 % suspension of CaO; N=3×10^4 W/m^3			
2.5	395/14	375/18	180/39
5.0	315/31	290/37	110/ 76
7.5	230/50	160/65	90/80
10.0	215/53	140/69	70/85
20 % suspension of CaO; N=3×10^4 W/m^3			
2.5	380/17	195/58	110/76
5.0	210/54	170/63	90/80
7.5	175/62	145/68	65/86
10.0	145/68	140/69	55/88

The degree of purification of wastewaters, treated in acoustic fields, grows by 1.3...1.5 times in comparison with untreated ones (only mechanical stirring) (Table 4). It was possible to significantly increase the degree of purification only at preliminary sonication with suspension of CaO. This made it possible to achieve the values of the CCO, which are permissible for dumping to the environment – 80 mg O_2/dm^3 (Table 5).

6. Discussion of results of examining the technology of wastewater treatment at meat processing enterprises

During non-reagent flotation of light-yellow juices, in the course of time we visually observed clearing of solution, change in its coloring and formation of dispersed elongated particles, which together with foam were carried out from the flotation plant. At the initial flotation stages, formation of dispersed particles was not visually observed. Subsequently, the solution acquired light-yellow coloring and became almost turbid. A change in the intensity of coloring at this flotation stage was accompanied by formation of finely dispersed particles, the coloring of which was virtually the same as that of original wastewater. Gradually, the particles were growing in size, became elongated, were more rapidly carried out with the bubbles to the surface, where they were accumulated as foam.

Within first 30 minutes, the degree of purification from organic compound did not exceed 15 %, and after one hour, it reached only 52 %. A slow change of the CCO at the initial stages of the process is caused by the fact that the process occurs through the stage of formation of dispersed particles due to decreased solubility of organic compounds in aqueous medium.

During non-reagent flotation of dark-brown wastewater, 50...70 % of the liquid phase were carried out from the flotation plant (depending on air consumption) within 10...20 minutes. Clearing of the liquid, remained in the flotation plant, was not visually observed.

The main reason for heavy foaming and, consequently, carrying out of water is very high alkalinity of wastewater of dark-brown coloring. Sodium and potassium salts of fatty acids, that is soap, contained in these wastewaters, are good solvents. That is why intensive foam is formed during flotation.

In addition, you must also take into account the fact that during flotation, the air with an unpleasant odor enters the atmosphere, which is not permissible for the enterprises that produce food products.

After adding solution of iron sulfate (II) (1 cm^3 of 20 % solution) to light-yellow wastewaters (150 cm^3), the partial oxidation of organic substances occurs. This is evidenced by the change of the ROP of the medium (Table 3). However, the change in the CCO during the stirring process is insignificant, this magnitude decreases only by 200...250 mgO_2/dm^3, that is by 10...15 % of the original value. This may be explained by two main reasons:

– in an alkaline medium the concentration of iron (II) ions in the solution is very low due to the formation of difficult-to-solve hydroxides of iron (II) and (III);

– low intensity of aeration of liquids during mechanical stirring.

In this regard, the following research was carried out at the intensive wastewater aeration and in presence of iron (II) sulfate.

After adding 10 cm^3 of solution of iron (II) sulfate to 250 cm^3 of wastewater of dark-brown coloring, pH practically did not change either after introduction of the coagulant or after finishing the experiment. This means that almost all iron ions transferred to the composition of difficultly soluble hydroxides. That is why hydroxides of iron (II) and (III) played mostly the role of coagulants, rather than the role of oxidation catalysts [7]. Indeed, during flotation, a certain amount of rather large aggregates that were carried out with foam was formed. It should be noted that heavy foaming was not observed.

In the liquid phase, which remained in the flotation plant, the formation of very finely dispersed particles was observed, and the liquid got somewhat lighter. Deposition of dispersed particles was very slow.

An hour later, the value of the CCO of the filtered solution was 2700 mg O_2/dm^3. After another hour, the value of the CCO decreased by ~500 mg O_2/dm^3. Respectively, efficiency of removal of organic compounds that cause a certain CCO value was 39 and 50 %.

After 25 cm^3 of solution of iron (II) sulfate were added to dark-brown wastewater (250 cm^3), the pH value decreased from 12.9 to 11.75, which is due to the acid nature of the coagulant. During the aeration, the pH magnitude decreased (Fig. 2) because of the oxidation of ions of iron (II) to iron (III).

An hour later, the value of the CCO decreased to 1190 mg O_2/dm^3, that is, efficiency of purification was equal to about 73 %. Within the following hour, the magnitude of the CCO decreased by another 400 mg O_2/dm^3 and the efficiency of purification increased by 82 %.

At pH magnitude of wastewater equal to about 7, and under other equal conditions, the value of CCO reached 1280 mg O_2/dm^3 within 60 minutes, the purification efficiency amounted to 71 %. The obtained results may be explained by the fact that with a decrease in alkalinity, the balance in the system of organic acid – organic salt shifted toward the acid, the stability of which is likely to grow. These considerations are proved by the fact that at pH equal to 5.1, the CCO magnitude equaled 1430 mg O_2/dm^3 (1 hour after the start of experiment).

Thus, it is necessary to note that in neutral and weakly acidic media, foaming was negligible, and solution clearing was more significant than in alkaline wastewater.

Dependence of the CCO magnitude of wastewater on pH during one-hour aeration is shown in Fig. 3.

The oxidation ability of substances-oxidants is known to increase with decreasing pH of the medium. Therefore, the following research was carried out under the terms of previous correction of the pH magnitude of wastewater to the acid area. To do this, we used diluted (1:3) sulfuric acid.

First, by using the solution of sulfuric acid, the pH of wastewater (200 cm^3) was decreased from 2.35 to 4.95. After adding 5 cm^3 of 20 % solution of iron (II) sulfate, the pH magnitude still decreased to 4.5.

It was found that addition of calcium oxide in the form of solid non-hydrated substance directly to wasterwater is less effective than applying solution of calcium hydroxide – lime milk. Though it might be expected that a hydroxide, which is characterised by a higher reactivity, will be formed during hydration of calcium oxide. In case of applying suspension of calcium hydroxide, hydrated calcium oxide and some amount of soluble calcium hydroxide, which is in equilibrium with non-soluble hydroxide, are introduced to wastewater. It provides a greater degree of binding of soluble organic compounds in the form of difficult-to-solve calcium compounds.

However, in case of applying the suspension of calcium hydroxide, a significant overconsumption of calcium hydroxide in the process of wastewater treatment is obvious. One of the known methods of intensification of processes involving solid phase reagents is the application of acoustic oscillations, in particular in the ultrasonic range [9]. Therefore, further studies were aimed at establishing efficiency of application of ultrasound radiation in the processes of wastewater treatment with calcium hydroxide. Results are shown in Table 4, 5.

The positive effect of application of acoustic oscillations may be explained as follows. Acoustic oscillations in various frequency ranges are known to create certain pulsing acoustic pressure on the elements of the heterogeneous system, which are different by nature and by properties [10]. First, during generation of acoustic oscillations, there occurs intensification of diffusion processes both in the entire reactive volume and directly near each solid particle of the heterogeneous systems. Second, ultrasound radiation excites the cavitation phenomenon which causes cumulative effect. It lies in the fact that during the closure of a cavitation bubble near a solid particle, microjets, directed to the surface of a solid particle, are formed near a solid particle. Velocity of cumulative jets reaches hundreds of m/s, which is why they have significant kinetic energy. As a result, the surface of particles of calcium hydroxide is destroyed with formation of numerous microparticles. As a result, the area of liquid and solid phases increases dramatically. This causes an increase in the rate of heterogeneous process. Besides, chemical interaction accelerates because newly formed particles are characterized by increased reactivity due to the excess of the surface energy on new – juvenile – surfaces.

Preliminary acoustic treatment of suspension of CaO was found to give a greater effect than sonication of the reactive medium, obtained by adding the reagent to wastewater. This can be explained by the fact that in the first case, the concentration of the CaO suspension is much higher than in the reactive medium after its adding to wastewater. That is why solid particles of CaO often get in the cavitation area, where their partial destruction and activation occur. As a result, the degree of CaO conversion in this case is higher. Therefore, it is necessary to activate the CaO suspension immediately before the introduction to wastewater.

Based on the performed research, the main stages of the purification process of wastewater of meat processing enterprises with the incomplete production cycle were selected. They include wastewater neutralizing, preliminary rough treatment on the bulk filter, flotation, reagent treatment, filtering, and disinfection.

The crucial stage of the process is reagent treatment of wastewater. It consists of alternate, with certain intervals, introduction of solutions of reagents: suspension of calcium hydroxide, activated by ultrasound, and solution of coagulant – iron (II) sulfate.

As a result of introduction of the 15…20 % suspension of calcium hydroxide, activated by ultrasound, protein substances, as well as water-soluble and emulgated fats and fatty acids, are deposited. To provide the maximum degree of using the reagent, its dosing is carried out by intensive stirring of the system, for example, using turbine or blade stirrers. After formation of flakes, solution of iron (II) sulphate (10 %) was added as a coagulant. Stirring was performed under laminar mode in order to avoid formation of iron (III) hydroxide, which negatively affects future filtering.

In both cases, wastewater is necessary to be neutralized and decontaminated after treatment. Decontamination is carried out with solutions of sodium hypochlorite, which allows us to simultaneously reduce the CCO magnitude.

7. Conclusions

1. Non-reagent flotation was found to decrease the CCO magnitude of wastewater of meat processing plants by nearly 50 %. Application of the coagulant – iron (II) sulphate – improves the effectiveness of purification to 80 %. However, the magnitude of CCO of treated wastewater exceeds the standard values that allow feeding them for additional purification by aerobic biological methods.

2. Application of the reagent – calcium hydroxide, activated by acoustic radiation of the ultrasonic range, enables us to achieve a residual value of CCO of less than 80 mg O_2/dm^3. It meets the standardizing indicators, at which it is allowed to discharge wastewater even into a natural water body.

3. For the treatment of highly concentrated wastewater of meat processing enterprises, we proposed the technology that consists of the following main stages: preliminary wastewater neutralizing, preliminary rough cleaning on the bulk filter, flotation, reagent treatment, filtering, and decontamination.

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