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## Electrochemical method of lead (II) ions removal from wastewater using granular graphite electrodes

The article presents an electrochemical method of wastewater treatment from Pb (II) ions with the help of granular graphite electrodes. The cathode was made of graphite particles and the anode was made of rod graphite. Features of wastewater treatment from heavy metals by using developed surface granular graphite electrodes were revealed. Electrochemical research was carried out in a flow mode in a two-chamber electrolyzer. Effect of different electrochemical parameters (current density, electrolyte flow rate, initial concentration of lead (II) ions in solution, size of graphite granules, and concentration of additional cations in solution) to the reduction process of Pb (II) on lump graphite was studied. Results reveal that the removal value of Pb (II) hit a peak at 150A/m<sup>2</sup> current density and gradually decreased at a higher value. The efficiency of electrolysis in the flow mode was demonstrated. It was found that the initial concentration of lead ions in the electrode process was insignificant, while the concentration of additional cations had a significant effect. Using very small particles of granular graphite electrodes caused an agglomeration. At an optimal condition, ( $i=150 \text{ A/m}^2$ ;  $V=150 \text{ ml/h}$ ;  $[\text{Pb}^{2+}] = 200 \text{ mg/l}$ ;  $s= 0.05 \text{ cm}^3$ ) treatment value of wastewater from Pb (II) ions reached  $97.6 \pm 0.3 \%$ .

**Keywords:** lead (II) ions, granular graphite electrodes, wastewater, heavy metals, flow mode, current output, removal degree, current density.

### Introduction

Rapid industrialization in the XXI century has led to problems such as heavy metal contaminated wastewater. Chemical industries are generating a large amount of wastewater contaminated with cadmium, zinc, lead and copper. These heavy metals, which have a density of more than  $5 \text{ g/cm}^3$ , are known to be toxic [1]. Wastewater pollutants can be classified by measure and chemical-physical properties. It is necessary to choose a suitable treatment method for each type of wastewater pollution. Wastewater treatment methods can be classified as chemical, mechanical, physico-mechanical and biological ones. They can be used together as a combined treating method [2]. Efficient methods of heavy metal removal from aqueous solutions include ion exchange, membrane filtration, coagulation, flotation, reverse osmosis, chemical precipitation, evaporation, solvent extraction, biosorption, adsorption, oxidation, electrochemical treatment, and others. Adsorption/ion exchange has been the most suitable method among these methods for heavy metal removal for great removal performance, cost-effectiveness and simplicity [3].

Our previous work represented a wastewater treatment by the ion exchange adsorption method. The research object was a model of wastewater containing  $\text{Zn}^{2+}$ ,  $\text{Pb}^{2+}$  and  $\text{Cd}^{2+}$  ions. A phosphorus-acidic cationite KRF-10P was used as a sorbent. Size of cationite, duration of interaction, and solution temperature were taken as affecting parameters and optimal condition was established. At an optimal condition, removal value of  $\text{Zn}^{2+}$ ,  $\text{Pb}^{2+}$ ,  $\text{Cd}^{2+}$  reached 96.1 %, 89 % and 91 %, respectively [4].

Industrial wastewaters produced while oil and gas production contain high concentrations of cadmium and lead ions. José E.D.V. Segundo and Aécio C.O. Feitoza developed an electrochemical reactor with a perforated steel cathode and expanded mesh three-dimensional DSA anode. Removal percentages were 96 % and 94 % for  $\text{Pb}^{2+}$  and  $\text{Cd}^{2+}$ , respectively [5]. Lead (II) ion adsorption of pristine graphite (6.13 % carbon, 93.87 % carbon) and oxidized graphite (15.97 % oxygen, 84.03 % carbon) was compared. The adsorption capacity of oxidized graphite was more than 70 %, while bare graphite showed only 41 % of adsorption capacity. This study results showed that graphite oxide was a suitable material for the  $\text{Pb}^{2+}$  absorption [6]. A hybrid biopolymer was used to extract Pb (II) ions from water. The hybrid biopolymer prepared by a simple

one-step galvanostatic polymerization consisted of poly(3,4-ethylenedioxythiophene)/ polystyrene sulfonate (PEDOT/PPS) and the biopolymer lignin (LG). The electrochemical results showed that the neutral solution of lead ions could be absorbed by applying a negative potential. LG can double the adsorption capacity of PEDOT/PSS from  $245 \text{ mg g}^{-1}$  to  $452 \text{ mg g}^{-1}$  [7].

Research on a new electrochemical way of lead (II) ions removal from industrial wastewater showed that iron and aluminum electrodes had a high level of lead, zinc and copper removal. At an initial concentration of  $50 \text{ mg/l}$ , and at optimal conditions, results showed 98.9 % of lead removal. But these electrodes have a short lifetime [8].

The electrochemical way of Pb (II) removal from wastewater by using iron and aluminum electrodes at low voltage was studied. The removal procedure included three processes i.e., chemical reduction, electrochemical reduction and electrocoagulation. The result showed that the removal value was directly proportional to the voltage and affected by the distance between electrodes and solution acidity. At pH value of 6, an applied voltage of 6V, and electrode distance of 2 cm, removal value of lead (II) was more than 90 % [9]. Electroplating effluents contain a high concentration of lead (II) ions. Researchers developed the removal method of lead (II) by electrocoagulation using iron as a sacrificial electrode. Removal experiment results showed that pH was an important parameter. Anode consumption increased with a decrease in pH while energy consumption increased with an increase in pH of the solution. As a result of the 90 min removal process, the removal value of Pb (II) reached 91.3 % [10].

The Taguchi method is a good technique for improving removal process performance, yield and productivity. It reduces manufacturing costs due to excessive variability in the process. Taguchi method contains several steps such as determining the quality characteristics (current efficiency, energy consumption), identification of the noise factors and test conditions, control parameters and their alternative levels, designing and conducting the matrix experiment, analyzing data, and determining the optimum levels [11]. A 3D reticulated vitreous carbon (RVC) cathode is used to remove lead (II) ions from aqueous solutions under acidic pH conditions. RVC cathode is coupled with a Zn anode. A batch electrochemical cell removed about 95 % of Pb (II) with a cell performance of 73 % [12]. Nanotechnology is also used for wastewater treatment. Yao Xing Lui and Jun Mei Yan used a stainless-steel net electrode coated with single-wall carbon nanotubes (SWCNTs@SSN). The mechanism of the electrochemical lead (II) removal method involved that Pb (II) ions were reduced and deposited on the surface of the SWCNTs@SSN cathode. An experiment was carried out under several parameters such as electrolysis time, pH value, applied voltage and initial lead concentration. After 90 min electrolysis, with an initial Pb (II) concentration of  $150 \text{ mg/l}$ , removal value reached up to 95 %. After electrolysis with the same conditions a good result was shown for other heavy metals [13].

As a result of literature review, it is obvious that electrochemical methods promise a high lead (II) removal degree. Granular graphite electrode (GGE) using electrochemical method is acceptable for many reasons. GGE has a high value of Pb (II) removal, the electrolysis method is simple, and it is cheap.

The purpose of the research is to study electrochemical methods of wastewater treatment from heavy metals using lump electrodes as a cathode in a flow mode, the influence of different electrochemical parameters to the removal degree (RD) and current output (CO) of metals. We examine an opportunity of using a lump electrode, composites metallic or graphite electrodes with large, developed surface, to avoid pointed out demerits. This treatment method is chemical-free and environmentally friendly, therefore, it could be a perspective direction and application of lumpy electrodes can intensify the process.

### *Experimental*

This work presents the results of the electrolysis of solution with an initial lead concentration of  $200 \text{ mg/l}$  on granular graphite electrodes. The necessary methods were chosen for carrying out the research. The research was conducted under direct current. A granular graphite electrode was taken as a cathode; a rod graphite electrode was taken as an anode.  $\text{Pb}(\text{NO}_3)_2$  was used to produce a model solution containing  $200 \text{ mg/l}$  of  $\text{Pb}^{2+}$  ions. Solution was acidified with 0.2 M of  $\text{HNO}_3$  acid. Electrolysis was carried out in a flow mode in a two-chamber electrolyzer (Figure 1). Solution containing lead (II) ions was passed at a certain rate through the granular electrode layer. Current density ( $i$ ), electrolyte flow rate ( $V$ ), initial concentration of Pb (II) in a solution ( $C$ ), size of graphite granule ( $s$ ) and concentration of additional cations in the solution were taken as the main parameters of electrolysis during the study to determine the features of removal degree (RD) and current output (CO) of  $\text{Pb}^{2+}$  ions in the solution. The duration of electrolysis ( $t$ ) and thickness of granular graphite layer ( $l$ ) was constant. An analyzer CTA-1 voltammeter was used for determining a low

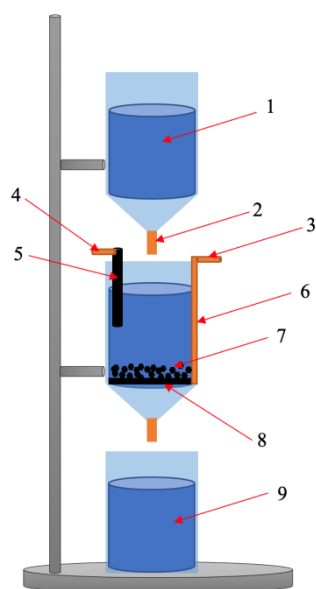
concentration of Pb (II) ions with very high accuracy. The process of determining the concentration of Pb (II) ions is shown in Figure 2.

The average mean of Pb (II) removal was determined by formula 1.

$$\bar{x} = \frac{\sum x_i}{n} \quad (1)$$

Formula 2 was used to determine a standard deviation:

$$\Delta x = \frac{\sum |x_i - \bar{x}|}{n} \quad (2)$$



1 — Pb (II) containing wastewater sample; 2 — water flow controller;  
3 — current input (-); 4 — current input (+); 5 — anode rod graphite;  
6 — copper wire; 7 — granular graphite particles;  
8 — cathode plate graphite; 9 — purified solution

Figure 1. Model of electrolyzer

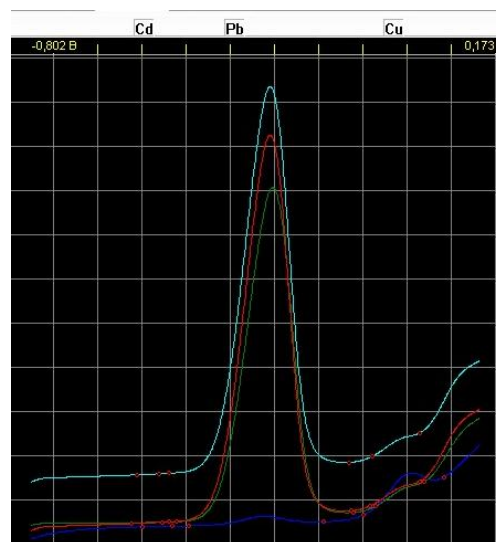
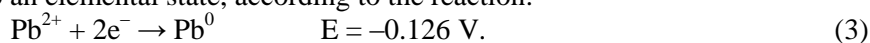


Figure 2. Determining Pb (II) concentration with a CTA-1 voltammeter

### Results and Discussion

The studies were based on the purification of wastewater from lead (II) ions by their electrolytic reduction on granular graphite cathode to an elemental state, according to the reaction:

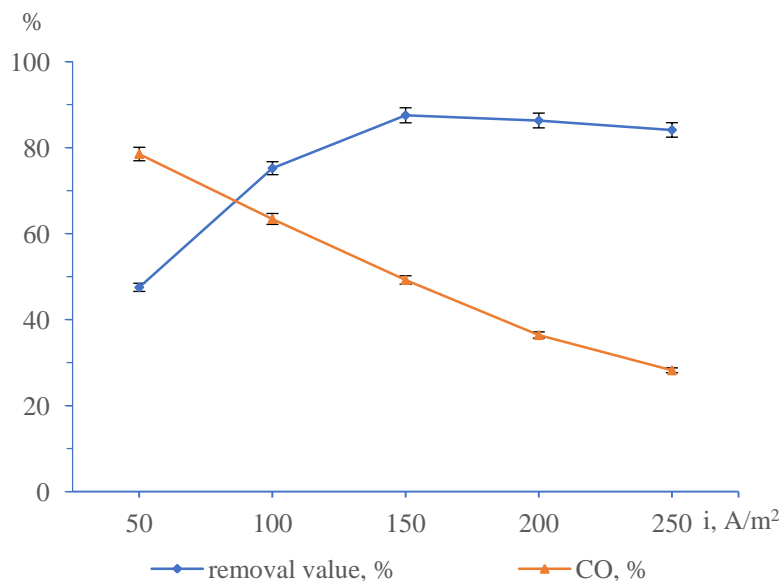


Discharge of water takes place at the anode:



Experiments were carried out between 50 and 250 A/m<sup>2</sup> current densities. It was observed that removal degrees grew steadily until the graphite electrode reached diffusion-limited current density (DLCD). Hydrogen emission occurred parallel to the lead removal at a high current density, consequently, removal of Pb (II) ions began to decrease (Fig. 3). The maximum degree of metal removal from the solution was achieved when the entire volume of the electrode operated at the limiting diffusion current mode. The decrease in the degree of lead extraction can be explained by the intensification of the competitive process of hydrogen evolution at high current densities.

The influence of electrolyte flow rate on lead (II) removal degree was determined. The range of electrolyte flow rate was between 50–350 ml/h. It was established that removal of lead (II) ions reached a maximum value at 150 ml/h electrolyte flow rate and decreased inversely proportional to the flow rate of electrolyte. When electrolyte flows at a high velocity, the reduction reaction of lead becomes less effective (Table 1). This phenomenon can be explained that at even higher flow rates, lead (II) ions cannot be properly reduced at the granular electrodes, and this leads to a decrease in the degree of metal extraction from the solution.



$[Pb^{2+}] = 200 \text{ mg/l}; s = 0.2 \text{ cm}^3; t = 0.5 \text{ h}; V = 0 \text{ ml/h}; l = 1 \text{ cm}$

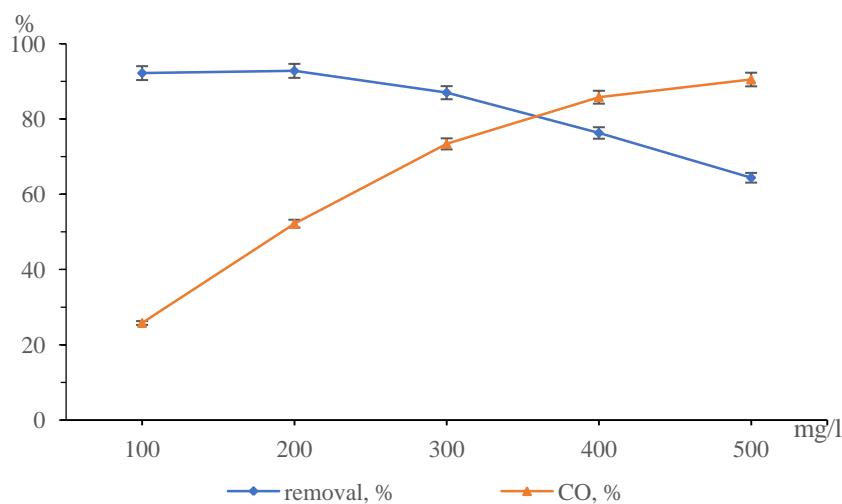
Figure 3. Effect of current density on Pb (II) ions removal degree

Table 1

**Effect of electrolyte flow on Pb (II) ions removal degree**  
*( $i = 150 \text{ A/m}^2; [Pb^{2+}] = 200 \text{ mg/l}; s = 0.2 \text{ cm}^3; t = 0.5 \text{ h}; l = 1 \text{ cm}$ )*

V, ml/h	50	100	150	250	350
$\alpha$ , %	89.2±1.5	91.7±1.3	92.8±1.5	86±1.0	80.5±1.2
CO, %	50.2±0.4	51.4±0.4	52.2±0.4	48.2±0.2	45.2±0.2

The removal value remains the same at a higher initial concentration of lead (II) ions. The rise of the initial concentration of Pb (II) ions causes saturation of solution and interferes with smooth polarization. It leads a process to occur in circumferential areas and the degree of Pb (II) ions removal gradually decreases. Current output gradually increases directly proportional to the initial Pb (II) concentration (Fig. 4).



$i = 150 \text{ A/m}^2; V = 150 \text{ ml/h}; s = 0.2 \text{ cm}^3; t = 0.5 \text{ h}; l = 1 \text{ cm}$

Figure 4. Effect of initial lead (II) ions concentration on Pb (II) removal degree

The influence of the electrode material on the electrolysis process is also considered. The maximum degree of extraction is observed on the graphite electrode (92.8 %), whereas on the lead electrode, this figure reached just under 68 %, as the copper electrode accounted for only 44 %. As a comparison, the experiment also was carried out without granular electrodes, which means only on a graphite substrate. It turned out that the degree of lead extraction didn't exceed even 15 %.

It was observed that change in graphite granule size affected the removal value of lead ions. Smaller granule size resulted in a higher removal degree and a higher current output while larger electrode granules resulted in a lower removal degree and less current output (Table 2). However, results of research carried out with the help of 1mm<sup>3</sup> granular graphite electrode showed that removal degree decreased steadily. It was found that small granules (< 0.01 cm<sup>3</sup>) agglomerated and became less effective because of the small reacting area.

Table 2

**Effect of graphite granular electrode size on Pb (II) ions removal degree**  
( $i = 150 \text{ A/m}^2$ ;  $V = 150 \text{ ml/h}$ ;  $[\text{Pb}^{2+}] = 200 \text{ mg/l}$ ;  $t = 0.5 \text{ h}$ ;  $l = 1 \text{ cm}$ )

s, cm <sup>3</sup>	0.25	0.2	0.15	0.1	0.05
$\alpha$ , %	89.9±0.7	92.8±0.8	94.4±0.8	96.7±0.6	97.6±0.3
CO, %	50.4±0.4	52.2±0.4	53±0.2	54.2±0.19	54.9±0.1

The influence of additional cations concentration in the solution on the Pb (II) ions removal degree and current output was determined during the electrolysis including copper ions. Lead (II) ions removal value and current output reduce gradually when copper concentration is increased in the solution (Table 3). This phenomenon can be explained by the reduction of copper ions parallel to the lead ions on the cathode.

Table 3

**Effect of additional cations concentration on Pb (II) ions removal degree**  
( $i = 150 \text{ A/m}^2$ ;  $V = 150 \text{ ml/h}$ ;  $[\text{Pb}^{2+}] = 200 \text{ mg/l}$ ;  $s = 0.05 \text{ cm}^3$ ;  $t = 0.5 \text{ h}$ ;  $l = 1 \text{ cm}$ )

[Cu <sup>2+</sup> ]	50	100	150	200	250
$\alpha$ , %	90.6±0.9	88±0.7	85.2±0.8	79.7±0.8	74.3±0.7
CO, %	51±0.25	49.4±0.2	47.8±0.2	44.8±0.22	41.8±0.2

At an optimal condition, ( $i = 150 \text{ A/m}^2$ ;  $V = 150 \text{ ml/h}$ ;  $[\text{Pb}^{2+}] = 200 \text{ mg/l}$ ;  $s = 0.05 \text{ cm}^3$ ) removal value and current output of Pb (II) reached 97.6±0.3 % and 54.9±0.1 %, respectively. It should be noted that if the solution is passed through several such kinds of electrolyzers, the removal value of Pb (II) ions will be higher. Deposited lead can be extracted from the electrode by burning the graphite.

### Conclusions

As a result of research, the influence of main factors affecting Pb (II) removal value was determined. It was observed that Pb (II) ions removal degree reached the maximum at 150 A/m<sup>2</sup> and decreased gradually because of hydrogen emission parallel to the lead (II) removal at higher current densities. At a high electrolyte flow rate, the reduction reaction of lead becomes less effective. A higher initial concentration of Pb (II) ions results in higher current output and lower removal value. The removal value and current output are inversely proportional to the size of cathode graphite particles, however, particles smaller than 0.01cm<sup>3</sup> become less effective because of particle agglomeration. Additional cations decreased the removal value and current output of Pb (II) ions. As a result of the research work the optimal conditions of electrolysis are established, at which ( $i = 150 \text{ A/m}^2$ ;  $V = 150 \text{ ml/h}$ ;  $[\text{Pb}^{2+}] = 200 \text{ mg/l}$ ;  $s = 0.05 \text{ cm}^3$ ) Pb (II) ions removal degree from wastewater accounted for 97.6±0.3 %.

The results of electrochemical research with the use of granular electrodes showed an opportunity of using electrochemical methods for wastewater treatment to intensify the electrode processes and to solve some ecological problems of the chemical and metallurgical industry. We consider that using granular electrodes is a new direction in the electrochemical branch on the solution of ecological problems.

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## Түйіршікті графит электродтарының көмегімен ақаба суды қорғасын (II) иондарынан тазалаудың электрохимиялық әдісі

Ақаба суларды құрамындағы ауыр металдардан тазалауда электрохимиялық әдіс өзге әдістерге қарағанда интенсивтілігі, тұрақтылығы және құрылғы конструкциясының қарапайымдылығы сияқты біршама қасиеттерімен ерекшеленеді. Мақалада түйіршікті графит электродтарын қолдану арқылы ақаба суларды Pb (II) иондарынан тазартудың электрохимиялық әдісі ұсынылды. Катод ретінде түйіршікті графит кесінділері, ал анод ретінде стерженді графит электроды қолданылды. Ақаба суларды ауыр металдар иондарынан дамыған беттік ауданды түйіршікті графит электродтарда электролиз тәсілімен тазарту ерекшелігі көрсетілді. Электрохимиялық зерттеу екі камералы электролизерде ағынды режимде жүргізілді. Түйіршікті электродтарда қорғасын (II) ионының тотықсыздану үрдісіне негізгі электрохимиялық параметрлердің (ток тығыздығы, электролит ағынының жылдамдығы, ерітіндідегі Pb (II) ионының бастапқы концентрациясы, графит түйіршіктерінің мөлшері және ерітіндідегі қосымша катиондардың концентрациясы) әсері зерттелді. Зерттеу нәтижесінде, ақаба сулардың қорғасын (II) иондарынан тазалану дәрежесі түйіршікті графит электродында 150 А/м<sup>2</sup> ток тығыздығында ең жоғары мәнге ие болатындығы анықталды. Электролизді ағынды режимде жүргізудің тиімділігі көрсетілді. Электродтық үрдіске қорғасын

иондарының бастапқы концентрациясының әсері елеусіз болса, қосымша катиондардың концентрациясы елеулі әсер ететіндігі анықталды. Түйіршікті графит электродтарының өте майда ұнтақтарын пайдаланғанда, оларға агломерация құбылысы тән екендігі көрсетілді. Электролиздің онтайлы жағдайларында ( $i = 150 \text{ A/m}^2$ ;  $V = 150 \text{ мл/сағ}$ ;  $[\text{Pb}^{2+}] = 200 \text{ мг/л}$ ;  $s = 0,05 \text{ см}^3$ ) ақаба сулардың Pb (II) иондарынан тазалану дәрежесі  $97,6 \pm 0,3 \%$  құрады.

*Кілт сөздері:* қорғасын (II) иондары, түйіршікті графит электродтары, ақаба сулар, ауыр металдар, ағынды режим, ток бойынша шығым, тазалану дәрежесі, ток тығыздығы.

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## Электрохимический способ очистки сточных вод от ионов свинца (II) с применением кусковых графитовых электродов

Электрохимический метод очистки сточных вод от тяжелых металлов отличается от других методов рядом свойств, таких как интенсивность, стабильность и простота конструкции устройства. В статье предложен электрохимический способ очистки сточных вод от ионов Pb (II) с помощью кусковых графитовых электродов. В качестве катода использовались куски гранулированного графита, в качестве анодов — стержневые графитовые электроды. Методом электролиза показана особенность очистки сточных вод от ионов тяжелых металлов на кусковых графитовых электродах с развитой поверхностью. Электрохимические исследования проводились в двухкамерном электролизере в проточном режиме. Изучено влияние основных электрохимических параметров (плотности тока, расхода электролита, начальной концентрации ионов свинца в растворе, размера гранул графита и концентрации дополнительных катионов в растворе) на восстановительный процесс ионов свинца на кусковых электродах. В результате исследования установлено, что степень очистки сточных вод от ионов свинца имеет максимальное значение при плотности тока  $150 \text{ A/m}^2$  на кусковом графитовом электроде. Показана эффективность электролиза в проточном режиме. Установлено, что начальная концентрация ионов свинца влияет на электродный процесс незначительно, в то же время существенное влияние оказывает концентрация дополнительных катионов. При использовании очень мелких кусковых графитовых электродов было показано, что для них характерно явление агломерации. При оптимальных условиях электролиза ( $i = 150 \text{ A/m}^2$ ;  $V = 150 \text{ мл/ч}$ ;  $[\text{Pb}^{2+}] = 200 \text{ мг/л}$ ;  $s = 0,05 \text{ см}^3$ ) степень очистки сточных вод от ионов Pb (II) составила  $97,6 \pm 0,3 \%$ .

*Ключевые слова:* ионы свинца (II), кусковые графитовые электроды, сточные воды, тяжелые металлы, проточный режим, выход по току, степень очистки, плотность тока.

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