Фармацевтичні відходи зі скла містять фармацевтичні речовини, які негативно впливають на навколишнє середовище та процес утилізації відходів. Для знешкодження таких речовин запропоновано запровадження способу електрохімічної деструкції. Додатково при цьому знезаражуються від бактерії Escherichia coli розчини фармацевтичних препаратів та підвищується екологічність утилізації відходів

Ключові слова: фармацевтичні відходи зі скла, навколишнє середовище, деструкція фармацевтичних речовин

Фармацевтические отходы из стекла содержат фармацевтические вещества, которые негативно влияют на окружающую среду и процесс утилизации отходов. Для обезвреживания таких веществ предложен способ електрохимической деструкции. Дополнительно при этом обеззараживаются от бактерии Escherichia coli растворы фармацевтических препаратов и увеличивается экологичность утилизации отходов

Ключевые слова: фармацевтические отходы из стекла, окружающая среда, деструкция фармацевтических веществ

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IMPLEMENTATION OF THE METHOD OF ELECTROCHEMICAL DESTRUCTION DURING DISPOSAL OF PHARMACEUTICAL GLASS WASTE

N. Samoilenko
PhD, Professor*
E-mail: samoilenko@kpi.kharkov.ua

I. Yermakovych
PhD, Assistant
Department of engineering of urban ecology
O. M. Beketov National University of Urban Economy in Kharkiv
Marshala Bazhanova str., 17, Kharkiv, Ukraine, 61002
E-mail: iryna.yermakovych@gmail.com

V. Bairachnyi
PhD, Professor*
E-mail: bairachniyvb@gmail.com

A. Baranova
Postgraduate Student*
E-mail: baranovaa647@gmail.com

*Department of chemical engineering and industrial ecology
National Technical University "Kharkiv Polytechnic Institute"
Kyrpychova str., 2, Kharkiv, Ukraine, 61002

1. Introduction

Formation and accumulation of pharmaceutical waste is becoming a global problem for society at present [1]. Development of the pharmaceutical industry and pharmaceutical sales market facilitates this. The volume of medicines' world sales may reach 1.5 trillion US dollars with an average annual growth of 5.9 % by 2021 according to forecasts [2].

Glass is widely used as packaging in the pharmaceutical industry. Glassware breakage is a subject of recycling and almost 100 % of glass can be reused according to EU [3] and Ukrainian legislation [4]. The use of glass waste in industrial processes reduces energy costs and reduces emissions of greenhouse gases at the same time.

Contaminated pharmaceutical glass waste (PGW) can be considered as a factor that negatively affects people's health, water and terrestrial ecosystems from environmental point of view. Pharmaceutical substances (PS) mainly contained in glass waste have the most harmful effect. Pollution of the environment with such pharmaceutical agents becomes an international problem and requires urgent solution.

In view of the above, it is an important scientific and practical task to introduce a method that would increase the ecological safety of PGW handling by destruction of pharmaceutical substances at the stage of waste collection and disposal.

2. Literature review and problem statement

Most of PGW are glass containers, they contain pharmaceutical substances that are usually easily released into the environment. European water bodies contain more than 3000 chemicals that relate to pharmaceutical products. The concentrations of some PS in natural waters of Europe, as well as the USA, range from $2.9 \cdot 10^{-6}$ g/l (Bisoprolol) to $0.025 \cdot 10^{-6}$ g/l (Ciprofloxacin) [5]. Development of advanced and complex analytical tools made it possible for scientists to register the concentration of PS in water to levels of particles per billion (ppb) and below in recent years [6].

It was determined that pharmaceuticals, which include many therapeutic groups, such as antibiotics, analgesics, anticancer drugs, contraceptives, and antidepressants, exhibit pronounced toxic effects to the environment [7]. The interaction of the components can occur and the toxicity of total exposure to organisms can be increased in a mixture which contains several pharmaceutical substances.

Pharmaceutical pollutants get into surface waters mostly because of the lack of effective biological treatment at urban facilities. This occurs due to two factors: low concentrations of pharmaceutical substances in waste waters and the impossibility of adaptation of silt microbiota to new medicines that appear on the medicine market [8]. This fact proves the expediency of prevention of getting pharmaceutical substances to municipal waste waters, including when handling pharmaceutical glass waste.

Ozonating, sorption, membrane purification, etc. are proposed to use for purification of water contaminated with pharmaceutical substances [9–11]. The use of powder activated carbon is recommended by authors [12] to increase the safety of drinking water from quinoline antibiotics.

The presence of diclofenac was noticed in purified waste waters and surface waters, as well as in drinking water. An ingress of diclofenac to the environment is a matter of particular concern in the world. It has harmful effect on terrestrial and aquatic organisms. In particular, it is proposed to use adsorption with cocoa husk to purify water solutions from diclofenac according to the latest research proposals [13].

The proposed methods of purification of water solutions from pharmaceutical contaminants are directed mainly at the purification of water from a separate pharmaceutical substance, but the issue of neutralization of different PS mixtures in a single process cannot be considered as sufficiently solved.

Pharmaceuticals have a negative impact on soil organisms and soil properties. Studies were conducted into such medicines as carbamazepine, diclofenac, fluoxetine and orlistat. They show that PS are included to the food chain of rainworms and change their internal chemical reactions [14].

The getting of PGW into landfills of household waste burial is regulatory permitted in some countries. The issue of prevention of it is less investigated. It is well known that glass waste decays for a very long time (500–1,000 years). It is likely that medical glass can decay longer than other types of glass waste, as it is resistant to the effects of chemical compounds. At the same time, studies show that the glass waste that gets to landfills alkalizes drainages considerably. It is recommended to store such waste under some roof to prevent atmospheric precipitation reaching them [15]. Glass also slows down the processes of microorganisms' vital activity at the surface layers of the soil. In addition, broken glass is a sharp object that also characterizes the danger of glass waste.

Organizations that have the appropriate technical base and special licenses are involved in transporting, destroying and disposing of waste in most countries. Recycling and disinfecting methods of medical waste, which contain PGW in its composition, in most cases have disadvantages. They relate to environmental factors (pollution of environmental elements, inefficient use of waste as a secondary material resource) [16–19]. And there are technical difficulties in addition. Medical glass gets to the burning furnace, melts and turns into a slag that sticks to the inner walls [20].

An analysis of the published data shows that the problem of PS environmental pollution was examined quite widely. The handling of PGW which contain pharmaceutical substances remains insufficiently investigated at the same time. First and foremost, it is necessary to include prevention of unacceptable environmental risk caused by PGW during waste collection and disposal to handling regulations.

3. The aim and objectives of the study

The aim of present study is to introduce a method of electrochemical destruction of pharmaceutical substances contained in PGW. Such method should enable to clear waste from solutions and residues of a medicine product, increase the resource value of waste and environmental safety of disposal technologies.

To achieve the set objective, the following tasks were defined:

– to substantiate theoretically and to confirm experimentally the expediency of electrochemical destruction of pharmaceutical substances contained in PGW;

– to carry out analytical studies which show the effectiveness of the process of degradation of pharmaceutical substances by the method of electrochemical destruction.

4. Study materials and methods

Electrochemical studies were carried out in solutions, which contained the following powdered pharmaceutical substances: diclofenac, β-estradiol, atenolol, furosemide, cefuroxime produced by Sigma-Aldrich (Belgium), of purity 99.9 %. NaCl or Na_2SO_4 (brand – Merck KGaA, Germany, chemical purity – 99.9 %) was added to the solutions studied. The electrochemical process was carried out in an open cylindrical glass cell. DC-Power Supply Laboratory device GPS-3030D was used as a current source. Electrodes: a graphite rod cathode, a platinum anode and ORTA anode. The degree of degradation of the substance in the samples under investigation was monitored by liquid chromatography with an ultraviolet detector (LC-UD) on Shimadzu HPLC device (Shimadzu Corp., Japan) and liquid chromatography with a mass spectrometer (LC-MS) of LC-MS Waters QTOF Xevo G2, Waters Acquity UPLC (Waters Corporation, USA) type.

5. Results of study on the introduction of electrochemical destruction method of environmentally safe handling of pharmaceutical glass waste

5. 1. Theoretical substantiation of expediency of destruction of pharmaceutical substances contained in PGW

Pharmaceutical waste (PW), by WHO definition, is the waste that contains medicines (medicines that have expired or no longer needed, objects that are contaminated with pharmaceuticals or contain such pharmaceuticals). On the other hand, PW are components of medical waste, which include expired, unused and contaminated medicinal products (MP) that require proper handling. In addition, pharmaceutical production waste is included to PW and also may contain MP. Taking into account the above, the determining characteristic of PGW is the presence of MP (pharmaceutical substances) in waste or contamination of glass containers by them.

The sources of the formation and accumulation of PGW are objects of the pharmaceutical industry, medical and health care institutions, pharmacies and pharmacy networks, veterinary establishments, population, research institutions and laboratories. Collection and disposal of waste is carried out by the subjects of economic activity, which have corresponding license.

Mineral resources (especially pure sands, kaolin, limestone, etc.) are consumed for production of medical glass, and glass waste is traditionally used as secondary material resources. Considering the above, it is important to provide conditions for the most effective and environmentally friendly technologies for the disposal of medical glass. The recycling of non-contaminated by pharmaceuticals PGW prevents harmful emissions to the environment and facilitates technological optimization of the process.

PGW feature is that the negative impact of waste has a combined effect on the environment. A generalized description of the negative impact of waste is shown in Fig. 1.

An analysis of the characteristics above shows that PS are the most health and ecological safety threatening, and also that they directly or indirectly negatively affect all elements of the environment. The worse effect is observed when PS gets to natural water. Liquid forms of MP contained in PGW are the main sources of such pollution in this case.

Taking into account all the facts mentioned, from the environmental point of view, the method of waste purification from the PS should ensure the neutralization of liquid MP to environmentally safe minerals.

Modern membrane purification technologies such as nano- and ultrafiltration, reverse osmosis cannot be widely recommended for the purification of PGW from pharmaceutical substances. This, first of all, is explained by insemination of waste that can occur in contact with sick people. Electrochemical advanced oxidation processes are the most effective for the removal of persistent organic pollutants, as well as pharmaceutical substances [22]. They include anodic oxidation, electro-Fenton and photoelectro-Fenton processes. Anodic oxidation is more rational in this case in comparison with other methods, since it does not require additional chemical reagents, or providing cathode with oxygen, or additional equipment.

Thus, it is advisable to use electrochemical destruction (anodic oxidation) for the removal of pharmaceutical substances extracted from PGW. Anodic oxidation makes it possible to mineralize organic matter, and thus, to reduce the negative impact of pharmaceutical substances on the environment.

5. 2. Method of electrochemical destruction of pharmaceutical substances contained in pharmaceutical glass waste

The studies were conducted with the most commonly used medicines: diclofenac, beta-estradiol, furosemide, atenolol, cefuroxime and their mixture. Diclofenac and beta-estradiol are priority pharmaceutical substances. They are included in the list of priority substances used in the monitoring of water bodies in EU and recommended by European Commission according to the Water Framework Directive (No. 2000/60/EU).

The concentration of PS in solutions was limited to 0.375 mg/l. The pH value of all prepared solutions was 6.65–7.25. The solution's temperature was 25 °C. NaCl or Na_2SO_4 (300–1000 mg/l) was added to the solution containing the pharmaceutical substance or a mixture of pharmaceutical substances in order to reduce the consumption of electric energy and increase the electrical conductivity of the solution.

The process was carried out using electrodes: a graphite rod cathode and a platinum wire anode (Pt), as well as a lamellar cathode of high-alloy steel and lattice ORTA (oxide-ruthenium-titanium anode) anode.

It was determined that the destruction of pharmaceutical substances proceeded with less efficiency in the solution containing Na_2SO_4 than in the solution containing NaCl 500 mg/l. Taking this fact into account, further research was conducted in the second medium.

Table 1 shows summary data on the modes of destruction of pharmaceutical substances by the method of anodic oxidation. The initial concentration of each pharmaceutical substance was 3 mg/l in all solutions. The process of anodic oxidation in solutions with different pharmaceutical substances and in a mixture of substances proceeded at a voltage of 31.5 V. The current strength was different.

It was established that PS destruction proceeds more slowly on platinum anode than on ORTA anode. Based on this, further research was carried out using the ORTA anode.

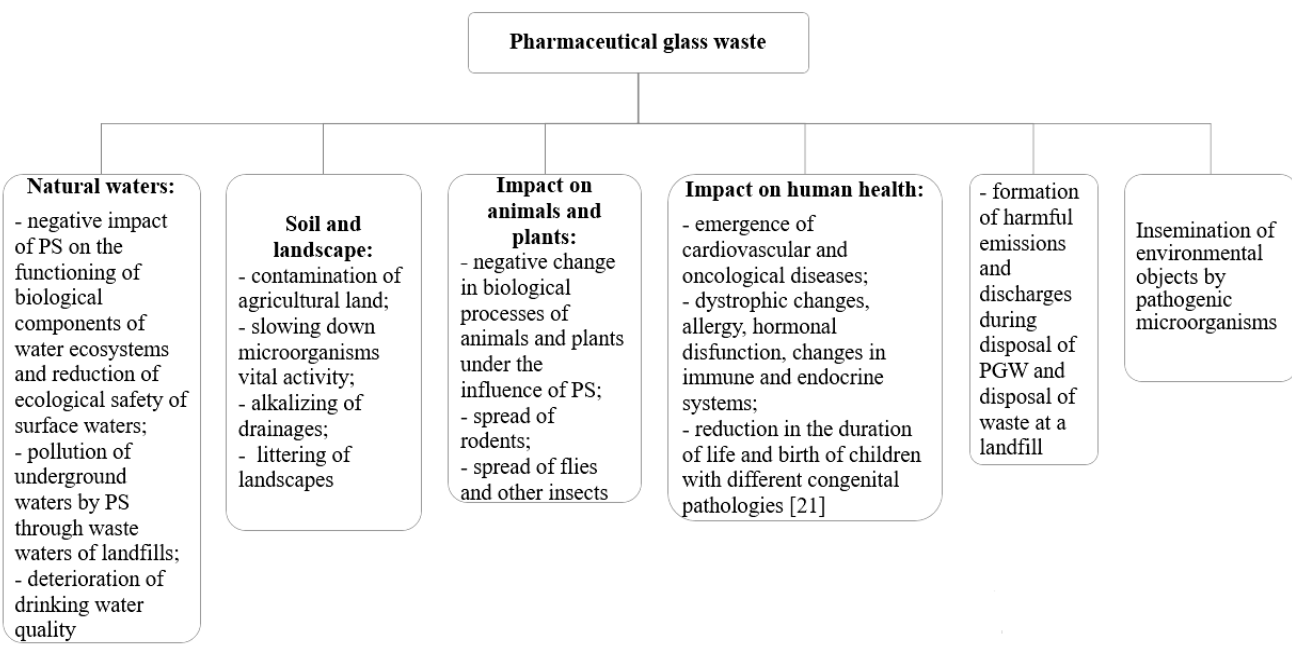

Fig. 1. Components of PGW negative impact on environmental elements and human health

Table 1

Parameters of PS destruction process by the method of anodic oxidation

PS name	Current strength (OPTA/Pt), A	Voltage (OPTA/Pt), V	Process duration (OPTA), min	Process duration (Pt), min
Diclofenac (DICL)	0.54/0.39	31.5	6	≤60
Beta-estradiol (B-EST)	0.59/0.35	31.5	1	≤6
Furosemide (FURD)	0.37/0.28	31.5	1	1
Atenolol (ATEN)	0.59/0.28	31.5	1	1
Cefuroxime (CEFX)	0.48/0.15	31.5	1	≤5
Mixture	0.55 (on OPTA anode)	31.5	10	–

The experimental data obtained on the destruction of substances using the ORTA anode are shown in Fig. 2

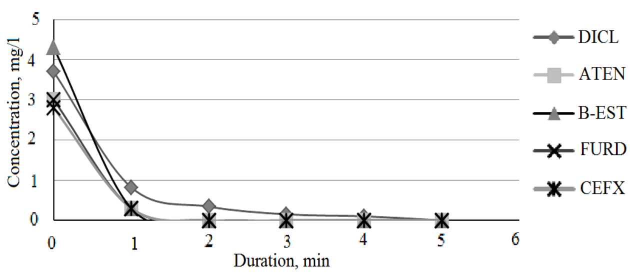

Fig. 2. Summary kinetic process of PS destruction with the use of ORTA anode. Concentration of NaCl – 500 mg/l

The degree of degradation of each pharmaceutical substance was analyzed with LC-UV and LC-MS analytical equipment and a mixture of five substances was investigated with LC-MS equipment only, as it is able to provide the most effective separation of complex mixtures.

The data from chromatographic analysis on determining the presence of priority substances used in the monitoring of water bodies in EU (diclofenac and beta-estradiol) in solutions after electrochemical degradation are given in Fig. 3, 4.

LC-MS chromatogram with UV detector of diclofenac is shown in Fig. 3. The wavelength was 254 nm. Duration of medicine availability in the sample was 6.77 min.

At a wavelength of 281 nm, the peak of beta-estradiol on the chromatogram was registered at minute 5.4 (Fig. 4).

Chromatographic analysis confirmed decomposition of diclofenac and beta-estradiol molecules in the process of anode destruction.

Fig. 5 shows LC-MS chromatogram with UV detector of a mixture of PS before the electrochemical destruction. Scanning of a mixture was carried out at a wavelength of 254 nm. Such wavelength is characteristic for the determination of substances with aromatic functional groups. The peaks of the chromatograms of these substances are well identified at this wavelength. This confirms the aromatic origin of PS contained in the mixture.

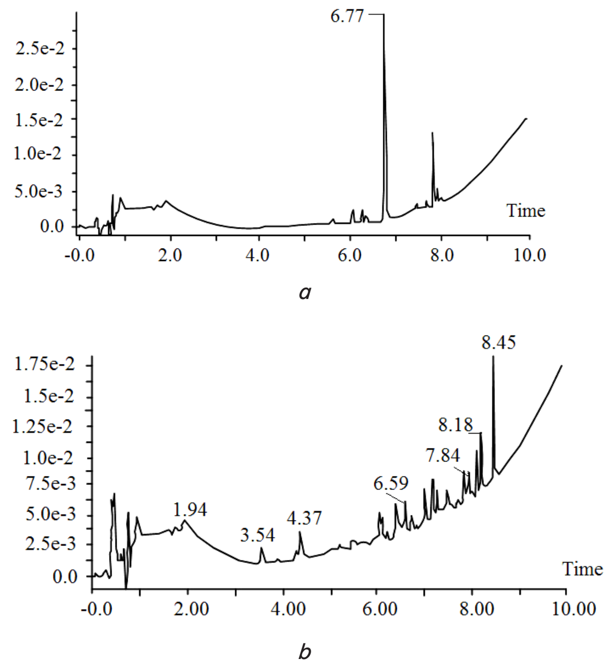

Fig. 3. LC-MS chromatogram with UV detector of diclofenac before (*a*) and after (*b*) conducting the electrochemical destruction

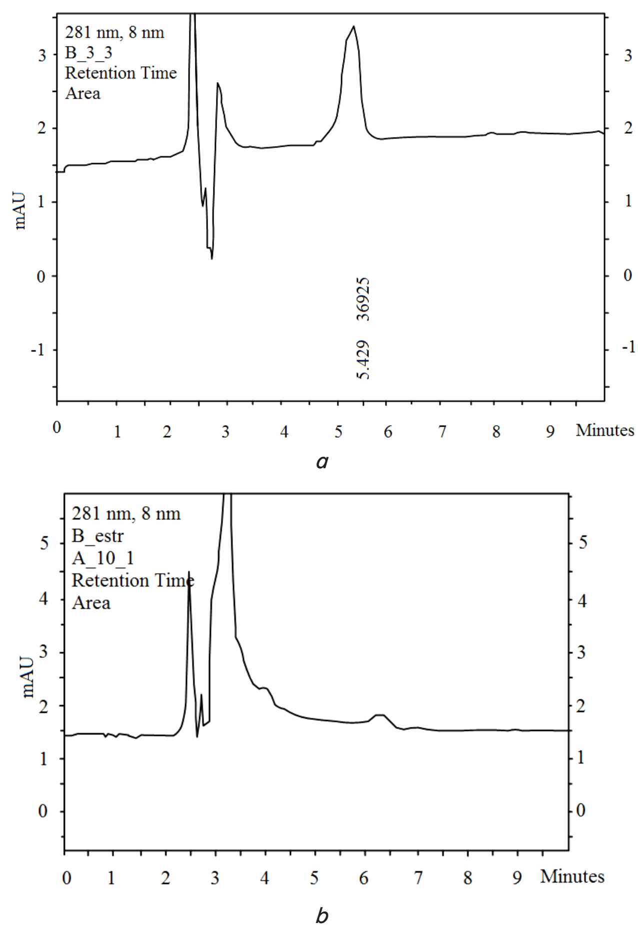

Fig. 4. LC-UV chromatogram of beta-estradiol before (*a*) and after (*b*) conduction of destruction

Duration of detection of each pharmaceutical substance was (min): atenolol – 0.78; cefuroxime – 4.02; furosemide – 4.98; diclofenac – 6.76.

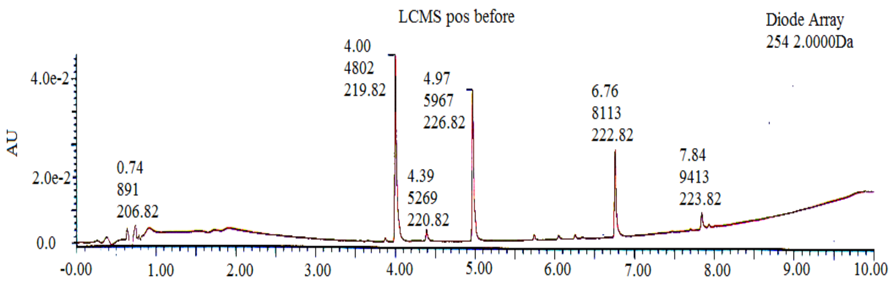

Fig. 5. LC-MS chromatograms with UV detector of the mixture before conducting the destruction

Destruction of the mixture of these priority PS on ORTA anode was carried out for 10 minutes. And after it was analyzed with LC-MS equipment at a wavelength of 254 nm.

Fig. 6 shows the result of scanning of a substances mixture after conduction of electrochemical oxidation. The data obtained show that all five pharmaceutical substances were not determined after destruction.

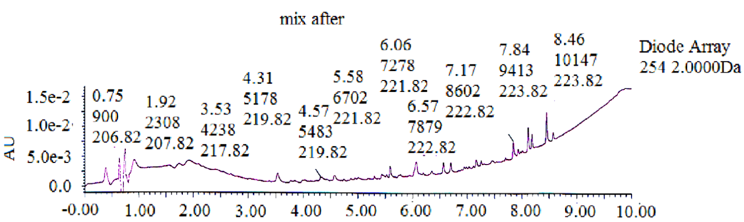

Fig. 6. LS-MS chromatograms with UV detector of a mixture after conducting the destruction

Thus, the analysis of a mixture of five pharmaceutical substances carried out with LS-MS equipment showed that all pharmaceutical molecules were completely destroyed during the process of electrochemical destruction.

6. Discussion of results of implementation of the method of electrochemical destruction of pharmaceutical substances contained in PGW

PGW's negative impact on the environment has complex character. It manifests itself in the contamination of environmental elements with pharmaceutical substances, glass and pathogenic microflora. The inflow of PS to water objects is the most widespread and dangerous, as it creates a threat to human health and negatively affects the biological components of surface water ecosystems and groundwater. In addition, pharmaceutical substances, such as diclofenac, beta-estradiol, etc., are resistant to biological degradation and are especially dangerous because of this fact.

Improvement of the environmental safety of PGW handling is possible by introducing of a method of electrochemical destruction of PS (EDPS) on waste treatment facilities. Studies conducted with the use of anodic oxidation showed complete destruction of diclofenac and beta-estradiol molecules, which were resistant to biochemical degradation, in solutions of these substances. Moreover, the process was more effective (less duration, higher degree of destruction) for diclofenac comparing with anodic oxidation, which was carried out using another anode and electrolytes [23].

The results of the studies showed the possibility of complete mineralization of a mixture of substances, which include diclofenac, beta-estradiol, furosemide, atenolol, cefuroxime. The lack of published data on the destruction of PS molecules under conditions of mixture does not make it possible to perform comparative analysis. But it may be noted that PS anodic oxidation parameters in a mixture did not differ significantly in relation to the parameters of destruction of individual PS. This fact makes it possible to suggest that such a pattern of relations may occur in the process of destruction of other PS mixtures from different pharmacological groups.

An additional effect that occurs during electrochemical destruction is complete inhibition of the development of a biotesting culture of *Escherichia coli* pathogenic family [24].

Implementation of EDPS method to the practice of PGW handling in medical institutions (for example, in a separate medical institution in a complex with an autonomous installation for the disposal of medical waste), pharmacy networks and pharmacies, as well as economic entities for the collection and disposal of wastes will significantly reduce environmental pollution with pharmaceutical substances.

Important results can also be obtained in the field of disposal of PGW. These results are related to extraction of PS from waste and getting a "clean" secondary material resource and include:

– assistance in the development and implementation of technology for disposal of medical glass, which has the greatest natural resource value;

– prevention of getting of PS to the products of waste recycling and disposal;

– improvement of technological conditions of PGW disposal process;

– making it impossible to form the waste of emissions and discharges, which are contaminated with pharmaceutical substances or derivatives of such substances, in the course of disposal. The studies conducted are the basis for further research on improvement of PGW handling. In particular, this concerns the treatment of pharmaceutical waste in the field of waste handling; creation of environmentally safe PGW disposal technologies that have the most resource-saving effect.

7. Conclusions

1. It was established that the getting of pharmaceutical substances contained in PGW into environmental elements creates the highest environmental risk. In this case, the pollution of natural water bodies by such substances is especially widespread and dangerous. Taking this into account, the priority direction of increase of the environmental safety of PGW handling is to prevent the flow of PS to sources of waste generation.

2. It was determined that increasing of PGW environmentally safe handling can be achieved by implementation of

EDPS method at the stage of waste collection and disposal. It has been experimentally proved that complete destruction of diclofenac, beta-estradiol, furosemide, atenolol, cefuroxime, and a mixture of these substances takes place in a solution with sodium chloride (500 mg/l) using the ORTA anode. The optimal modes and conditions to carry out the anodic oxidation process for each of these five PS and their mixtures were determined.

3. It was noted that the process of PS electrochemical destruction can be accompanied by decontamination of solutions of pharmaceuticals from *Escherichia coli* bacteria. The introduction of EDFR method contributes to the improvement of the quality of PGW as a secondary material resource and to the improvement of technological processes of pharmaceutical waste disposal.

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