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TREATMENT OF LEACHATE BY THE CARBON MATERIAL OBTAINED FROM POLYMER FRACTION OF MUNICIPAL SOLID WASTE

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Abstract: *In the study, the process of anaerobic thermal processing of polymer fraction of municipal solid waste using a horizontal and vertical type of reactor has been developed in order to obtain a new carbon sorbent. The productivity of carbon material by the horizontal method is 20%, while vertical type of reactor is about 30%. The sorption potential of some heavy metals and microbiological indices from leachate water of Tbilisi municipal solid waste polygon has been studied. According to estimates, the sorption potential of pollutans from wastewater using our new sorbent is average 60-98%.*
Keywords: *carbon materials, sorbent, thermochemical processing, leachate, treatment, heavy metal, microbiological indices.*
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Introduction

In developing countries, including Georgia, the main part of the solid waste generated throughout the country is placed in the so-called - open landfills. This type of waste disposal leads to the pollution of environmental components (soil, water, air), which in turn affects human health, for mostly it does not comply with the principles of sustainable waste management and circular economy protocols.

Nowadays, improving solid waste management (MSW) is one of our country's priorities aimed at harmonizing the development process of waste management of European and the world's waste management policies. At present, about 50 official landfills of open type are registered in Georgia, the largest of which serves the population of the capital city [1,2].

Currently, the main part of the MSW generated throughout the country is placed in the so-called - open landfills (90%). The largest official landfill (Norio SWDS) in Georgia serves the capital city - Tbilisi, and is located to the South-East of Tbilisi, at the east of the settlement (population - 2500 persons and 30 km from capital city) [3,4,5].

Operating of the landfill started on November 15, 2010. The body of the polygon of MSW consists of the cells, in which solid waste dumped by means of a bulldozer and loader is distributed and compressed with special compactors and then cover by local soil. In the process of work, the waste is moisturizing with existing leachate of the landfill. When the landfill cells are filled, each cell is covered with an insulating coating containing a special geo-membrane and the surface of the closed ones are re-cultivated. Landfill gas is collected through shafts, which are set as soon as the cell is arranged, and whose main function is to prevent methane ignition (landfill gas is not used it is emitted in to the atmospheric air).

The reverse osmosis device is provided for treatment the leachate waters but this device out of service for over 4 years and the leachate water cannot be treated in accordance with the legal norms for discharge into the sewerage system [6].

To solve this problem, the leachate water from sedimentation system is pumped to the surface of the working waste storage cell by using pumping machines, therefore it circulates: from sedimentation system to the body of the landfill - compressed waste. This activity moistens the waste and, consequently, activates the biodegradation process, and also, significantly increases the degree of pollution of leachate water. In addition to all of the above, there is a high risk of environmental pollution, which is mainly due to the high probability of emergent discharge of the leachate water, namely: in the case of heavy precipitation, water bodies (sedimentation systems) are the most likely to overflow and discharge contaminated water into the collector and then to the river Norikhevi, that flow in the natural ravine nearby, and then in Lochini, and finally to the trans-boundary river Mtkvari, the main river of the capital city.

Moreover, one of the main problems is the increased content of the toxic and cancerogenic pollutants in the leachate water, which are formed because of migration due to the disposal of mixed waste.

Over the years in Georgia MSW volume is growing about 1.7% a year [7]. From 2015 till 2017 on the basis of the Institute of Hydrometeorology of Georgian Technical University, we carried out the project "Elaboration of Methodology for Determination of Accumulated Amount and Morphological Composition of Municipal Solid Waste in Georgia and Database Creation." The project was funded by the National Foundation, within which the morphological composition of municipal solid waste generated in the country was determined (Fig. 1) [4]. The results show that a share of one of the most difficult to decompose fractions - plastic is 14%, namely 126.000 tons / year. The goal of our research was to minimize plastic waste, which includes the use of the method for rational and environmentally beneficial minimization of municipal solid waste, developed at the R. Agladze Institute of Inorganic Chemistry and Electrochemistry of Ivane Javakhishvili Tbilisi State University. The methodology was developed on the basis of the method of obtaining of carbon materials from cellulose-containing waste, patented by the Institute [8,9,10]. In particular, low-temperature thermochemical processing of such hardly decomposable fractions of MSW as polymeric waste, in order to obtain carbon and study of the possibility of its use as a sorbent for heavy metals and microbiological substances from wastewater.

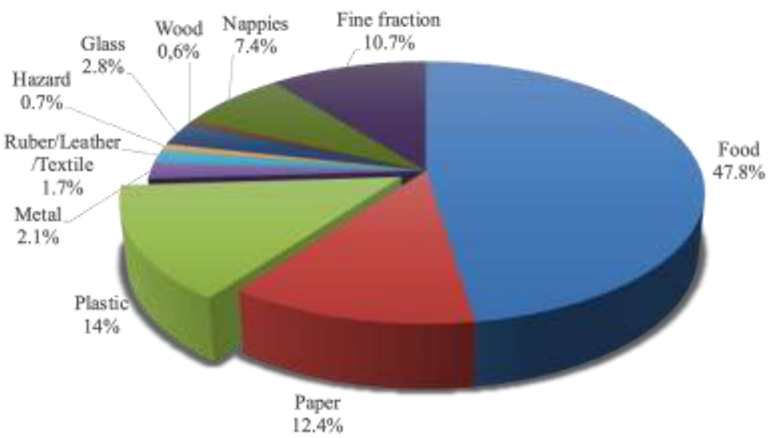


Fig. 1: Morphological composition of MSW [3,4,7]

For minimization of MSW volume and to solve the problem of leachate pollution, the aim of our research was to obtain a carbon sorbent from a difficult to decompose and most harmful

MSW fraction - polymer fraction of MSW and use it for treatment the leachate generated at the SWDS.

Experimental Part

Our research started since November 2021. The first stage of our research was the development of the rational method of obtaining carbon materials from polypropylene fraction of MSW. As part of our research, in order to obtain carbon material, initially, the polypropylene from polymer fraction of MSW was separated and crushed for further thermochemical processing to obtain carbon materials.

Thermochemical processing mode was selected based on the results of thermogravimetric analysis (STA 2500 Regulus. Simultaneous Thermal Analysis) [11]. Figure 2 shows the thermal gravimetry (TG) and differential thermal gravimetry (TGA) curves. TG describes the change in the mass of a sample depending on the increase in temperature, and TGA is the derivative of TG signal in the time, i.e. the rate of change in the mass of the sample, which allows establishing the moment at which the change in mass occurs most quickly. The peaks indicate characteristic temperatures of decomposition. The thermogravimetric method is effective on condition that the sample releases volatile substances and, accordingly, thermogravimetric analysis is widely used to determine the degradation temperature of polymers.

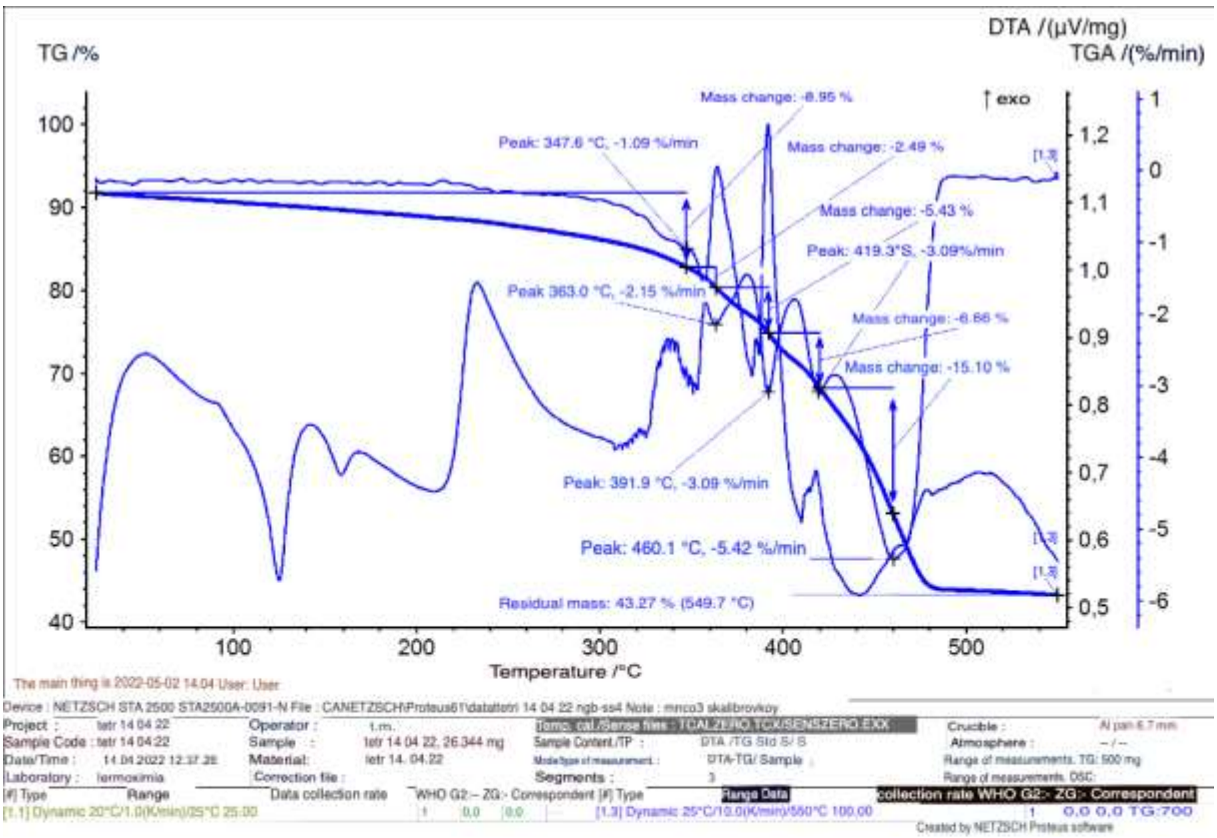


Fig. 2. Thermogravimetric and differential thermal analysis data (Polypropylene)

The TG curve shows that the thermal decomposition of the sample can be divided into three stages: 1) <300°C, 2) 300-450°C, 3) >450°C. At the first stage, the mass loss is insignificant, which can be caused by the evaporation of water and the decomposition of trace amounts of organic substances; at the second stage, the mass of the sample decreased significantly (~45%), which complies with the TGA curve with a peak of mass loss at a temperature of 300-450°C, which can be explained by the decomposition with removing of volatile substances. As can be seen in the figure, the curve has only one sharp peak in the temperature range of 300-450°C and it is obvious that the volatile substance appears precisely in this range. In the third stage, the trend of mass loss slows down, which means that the solid residue almost does not decompose.

The optimum temperature of thermochemical treatment of polymer waste samples was determined as 420 - 450°C based on abovementioned thermogravimetric study.

In order to obtain carbon materials from polymer waste, a stainless-steel horizontal type reactor was constructed, which is a hermetic capsule with two stainless steel pipes. The sample was placed in the reactor, in which anaerobic conditions were achieved by nitrogen flow delivered through the stainless-steel pipe [12].

During the degradation of plastic by heating at the temperatures 420-450°C, macromolecules break down into smaller fragments consisting of mixtures of hydrocarbons in the form of gas, liquid and solid. Thus, when the amount of heat supplied exceeds the dissociation energy of the various bonds, they break. Since the chain C-C bonds have the similar strength, the main chain of the polymer is broken randomly. This type of rupture is typical for polypropylene (PP), whereby shorter chains of hydrocarbons are formed by a free radical mechanism. High temperature leads to the formation of a volatile fraction, which can be further divided into condensable liquid fuels, non-condensable hydrocarbon gases, and also solid fractions. The yield of products and their ratio depends on the qualitative composition of the raw materials, temperature and time conditions, as well as the type of reactor.

As the temperature gradually rose, gases (hydrocarbons) began to be released from the second tube of the reactor, and a liquid organic fraction was formed at a temperature of 350 °C, which stopped being released when it reached 420-450°C. The process was continued approx. 2 hours, after cooling the reactor was opened and the obtained carbon materials were weighed.

The results of thermal treatment of polymer waste by means of our horizontal reactor show that the yield of carbon obtained from 200 g polypropylene waste using the mentioned thermochemical method is up to 20%, the remaining fractions are gaseous and liquid products of thermochemical treatment about 80% [12].

To increase the productivity of carbon material and reduce the by-products of the chemical-thermal treatment of polymeric waste samples (polypropylene), a vertical-type reactor was constructed. When using a vertical-type reactor, the temperature of obtaining of the carbon is 450°C, the product yield is approx. 30% [13].

The next step was the study of physical characteristics of obtained carbon material for evaluation of sorption capability towards potential pollutant of ecosystems. One of the most important indices of carbon sorbents is their porous structure. Carbon sorbents have a complex porous structure; they belong to the group of heterogeneous porous materials [14, 15, 16]. To solve the tasks, we studied the surface area and porosity of carbon samples obtained by horizontal and vertical methods from polypropylene waste, by using:

1. For surface morphology – SEM: Hitachi TM 3000;
2. For surface area and porosity– Micromeritics: Gemini VII Version 2390T.

The surface morphology of was examined using a Scanning Electronic Microscope equipped with an Energy-Dispersive X-ray analyzing system. The point is about the rough surface with some smooth particles with several cracks having spongy pores. This confirms that material contains mesopores. The cracked structure with pores confirms the amorphous structure of the formed chars. Its surface was heterogeneous indicating the presence of pores of various shapes and sizes.

Figure 3 shows micrographs of the surface of sorbents with magnification. During relatively high-temperature processes, a partial melting of the organic matter of the polymer occurs with the release of volatile fractions, which causes a round and oval shape of the pores. Dark spots associated with pores near the surface are also visible on the surface.

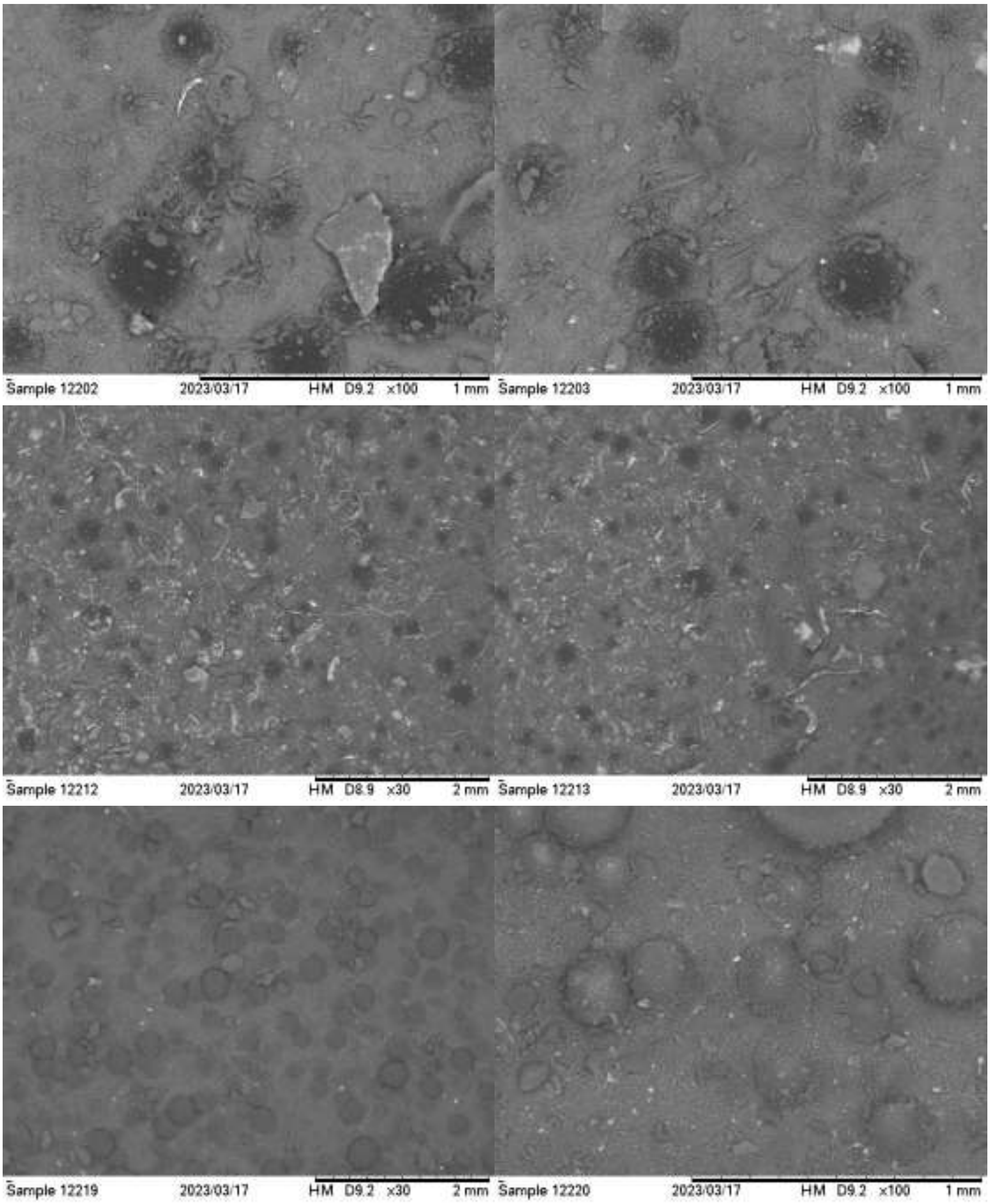


Fig. 3. Micrographs of the surface of carbon sorbents

The textural properties - surface area and porosity of a sorbent are important factors in determining its capacity for sorption.

We started with sizing of particle of mixed carbon materials by Mastersizer 2000. Result of analysis shows, that the particle sizes of mixed carbon materials were ranged from 0.02µm to 2.000 µm, with maximal density on 832 µm particle size (Fig. 4).

To homogenize the particle size, the carbon materials was processed into micro-ground. To achieve better sorption capacity the textural characteristics of carbon samples before and after micro-ground were studied. Surface areas and porosity of carbon material from polymer (polypropylene) were specified using the Brunauer-Emmett-Teller (BET) and Langmuir methods based on adsorption of isotherms of non-reactive nitrogen [17].

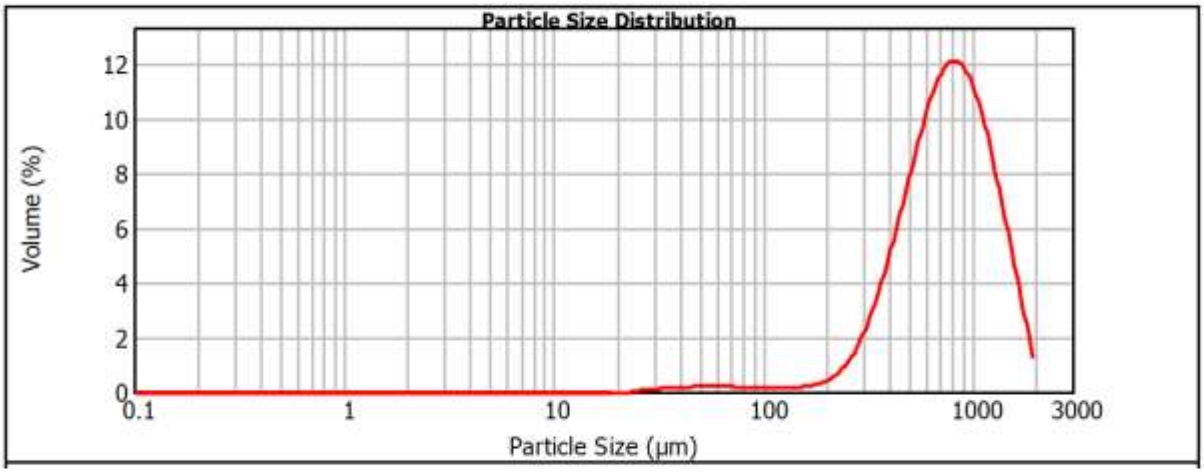


Fig. 4. Particle size of mixed carbon materials

In this study, we evaluated the BET and Langmuir surface areas of carbon material obtained by horizontal and vertical low-temperature chemical process. Results of analysis before micro-ground and after micro-ground are show in Table 1. S_{BET} and $S_{Langmuir}$ was determined from adsorption isotherms in the range of relative equilibrium pressures of nitrogen vapor P/P_0 = 0.05–0.3. Nitrogen adsorption–desorption isotherms (T_{ads} = 77.4 K) were obtained on Porometer Gemini Micromeritics.

Table 1. Surface area and porosity of obtaned cabon material (from polypropylene)

Parameter	Unit	Carbon Material Sample (from polypropylene)	
		before processing	after processing
Sample density	g/cm^3	0.510	0.626
BET surface area	m^2/g	4.4472	11.1843
t-plot micropore area	m^2/g	1.16854	4.6596
t-plot external surface area	m^2/g	2.7618	6.5247
t-plot micropore volume	cm^3/g	0.000807	0.001837
Max. pore volume	cm^3/g	0.001976	0.003975
Median pore width	nm	1.4473	1.4545

Table 1 shows that as a result of mechanical processing (grinding) of the obtained carbon material, favorable parameters for sorption improved.

From July 2022 till January 2023 every month, we were sampling of leachate water from the storage tank of the leachate water on the territory of MSW landfill of Norio. Physical and chemical indices were measured in the field, and then in the laboratory, the determination of heavy metals (Pb, Cr, Cd, Hg) and microbiological indices (Total coliforms, E. coli) were carried out according to ISO methods [18-21].

The study of the sorption of selected pollutants using the sorbent obtained by us, was carried out as follows:

To 50 ml of the studied water was added 1 and 2 g of carbon materials (unprocessed sorbent) obtained by vertical type of reactor from polymeric waste, and held for 24/72 hours. The samples were then filtered through a filter (pore size 2-3µm). In the obtained solutions Pb, Cr, Cd, Hg, Total coliforms and E. coli were established by the abovementioned methods.

Results and discussion

Results of determination of heavy metals and microbiological indices in samples of the studied leachate water showed in Table 2.

Table 2. Content of some heavy metals and microbiological indices in leachate water of Tbilisi SWDS (with Maximum Permissible Discharge (MPD), according to Georgian legislation [22])

Metals/ Indices	tºC	Cr	Cd	Hg	Pb	Total Coliforms	E. Coli
July. 2022	32.1	1.79	0.77	0.51	1.87	559 950	<500
August. 2022	33.8	2.11	0.87	0.61	2.13	28 250	<500
September. 2022	20.2	1.06	0.52	0.38	1.13	71 500	500
October. 2022	12.8	2.21	0.79	0.50	2.40	3 750	<100
November. 2022	9.8	2.36	0.87	0.60	2.25	4 650	<100
December. 2022	4.3	2.59	1.07	0.76	2.20	5 950	<100
January. 2023	1.8	2.86	1.37	0.90	2.35	3 907	0
Average	16.4	1.65	0.72	0.5	1.71	-	<500
MPD Wastewater	-	1	1	0.5	1	5000	-

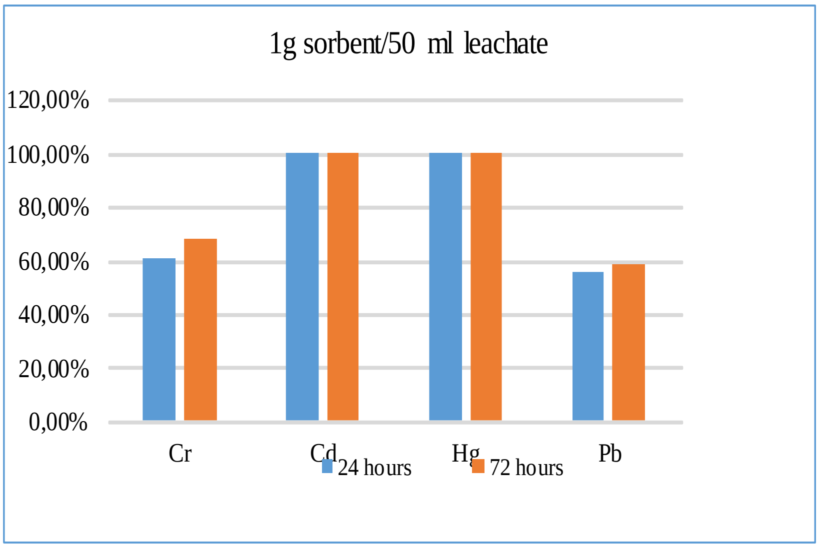
The microbiological analysis of leachate water shows that the average concentrations of Total Coliforms in research waters have maximum levels during the summer season (excess of MPD of Total Coliforms in 44 times) and minimum values in winter (near threshold of MPD). The physical-chemical analysis of leachate water is indicative that the concentration of heavy metals exceeds 1.5-2 times the MPD of wastewater and in case the leachate water ingress into the environmental objects can heavily impact.

The study of sorption potential of carbon sorbents obtained from polymer (polypropylene) fraction of MSW indicates that they were characterized by good sorption capacity to the analyzed heavy metals and microbiological contaminants of wastewater. In particular, on average, the sorption of metals is 81%, with a maximum for Cd and a minimum for Pb (Table 3), which most likely results from their initial contents in the studied leachate water samples (Table 2).

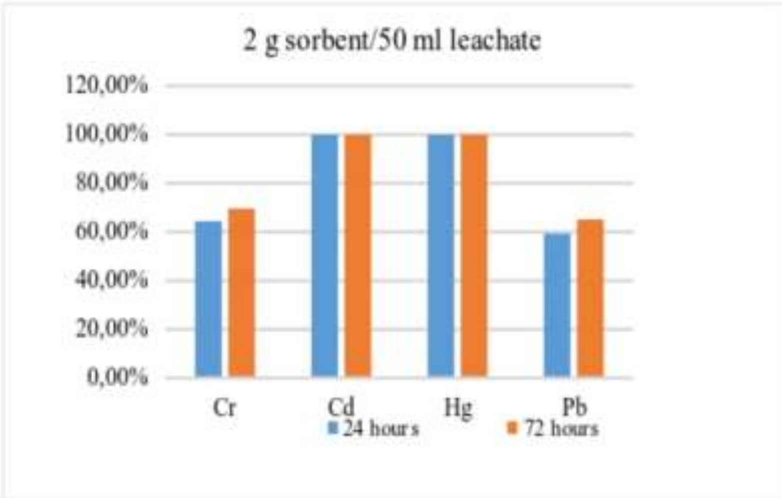
Table 3. Content of some heavy metals in leachate water of Tbilisi SWDS after treatment by carbon sorbent from polypropylene waste fraction (sample 50 ml)

Metals/ Indices	Average (VII/2022-I/2023) mg/l	Sorption %				Average after sorption, mg/l	MPD Wastewater, mg/l
		1 g carbon material per 50 ml Leachate		2 g carbon material per 50 ml Leachate			
		24 hours	72 hours	24 hours	72 hours		
Cr	2.14	60.76	67.71	64.26	69.17	0.74	1.00
Cd	0.89	99.99	99.99	99.99	99.99	<0.01	1.00
Hg	0.61	99.93	99.94	99.94	99.94	<0.01	0.5
Pb	2.05	55.59	58.67	58.81	64.94	0.84	1.00

Based on the Table 3, we can conclude that the optimal degree of sorption, in 80% of cases, are achieved within 24 hours and in the case of using 1 g of sorbent for 50 ml of the studied leachate water (Fig. 4, parts I and II).



(I)



(II)

Fig. 4. (I and II) Sorption capacity of studied heavy metals from leachate depending on the time.

The sorption of Total Coliforms is on average 75% and, as a result, the average value of its decreases to MPD (Table 2 and Table 4). The same as in the case of heavy metals are achieved within 24 hours and in the case of using 1 g of sorbent for 50ml of studied leachate water samples (table 4).

Table 4. Content of Total Coliforms in leachate water of Tbilisi SWDS after treatment by new carbon sorbent (MPD– 5000 MPN/100ml)

Total Coliforms	MPN/100ml	Sorption %				After sorption, MPN/100ml			
		1 g carbon material per 50 ml Leachate		2 g carbon material per 50 ml Leachate		1 g carbon material per 50 ml Leachate		2 g carbon material per 50 ml Leachate	
		24 hours	72 hours	24 hours	72 hours	24 hours	72 hours	24 hours	72 hours

September, 2022	71 500	72.87	73.29	-		19400	19100	-	-
October, 2022	3 750	69.79	73.33	-	-	1133	1000	-	-
November, 2022	4 650	72.04	72.58	-	-	1300	1275	-	-
December, 2022	5 950	-	-	87.41	87.78	-	-	749	727
January, 2023	3 907	-	-	70.92	71.33	-	-	1136	1120

Therefore, the degree of sorption of studied pollutants varies from 55.59% to 99.99% (Table 3 and 4).

In addition, treatment of leachate water with the sorbent significantly reduces its organoleptic indices (odor, color), Fig. 5.

Fig. 5. Tbilisi landfill leachate water before treat (right) and after treat (left) by means of mixed carbon sorbent from polypropylene MSW fraction (1g sorbent/50ml leachate, 24 hours under static conditions).

Conclusions

The research proves that the use of sorbent obtained from polymeric waste can be the effective and cheap method of leachate treatment from studied pollutants. The use of the carbon sorbent reduces the heavy metals and microbiological indices (to much lower level than MPD) and in case of emergency flooding, leachate will not have a dangerous impact on the ecosystems.

And most importantly, for the development of the circular economy, it is possible to usefully minimize polymer waste for mitigation of its environmental impact, with the use of the method of vertical thermochemical processing of MSW polymeric fraction, developed within the frame of our study, because of the complete processing of the liquid by-product and the return of the released gases (and their use as fuel for) to the process.

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FİLTRAT SUYUNUN TƏMİZLƏNMƏSİ ÜÇÜN BƏRK MƏİŞƏT TULLANTILARIN POLİMER FRAKSİYASINDAN ALINAN KARBON MATERİALLARININ SİNTEZİ

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Xülasə: Tədqiqatda yeni karbon sorbentinin alınması üçün üfüqi və şaquli reaktorlardan istifadə etməklə bərk məişət tullantılarının polimer fraksiyasının anaerob istilik emalı prosesi işlənib hazırlanmışdır. Üfüqi üsulla karbon materialının məhsuldarlığı 20%, şaquli tipli reaktorla isə təxminən 30% təşkil edir. Tbilisi bərk məişət tullantıları poliqonunun filtrat sularından bəzi ağır metalların sorbsiya potensialı və mikrobioloji göstəriciləri öyrənilmişdir. Hesablamalara görə, yeni sorbentdən istifadə zamanı çirkab sulardan çirkləndiricilərin sorbsiya potensialı orta hesabla 60-98% təşkil edir.

Açar sözlər: karbon materialları, sorbent, termokimyəvi emal, filtrat, təmizləmə, ağır metallar, mikrobioloji göstəricilər.

СИНТЕЗ УГЛЕРОДНЫХ МАТЕРИАЛОВ, ПОЛУЧЕННЫХ ИЗ ПОЛИМЕРНОЙ ФРАКЦИИ ТВЕРДЫХ БЫТОВЫХ ОТХОДОВ ДЛЯ ОЧИСТКИ ФИЛЬТРАТНЫХ ВОД

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Аннотация: В исследовании разработан процесс анаэробной термической переработки полимерной фракции твердых бытовых отходов с использованием горизонтального и вертикального типа реактора с целью получения нового углеродного сорбента. Производительность углеродного материала при горизонтальном способе составляет 20%, а при вертикальном типе реактора - около 30%. Исследован сорбционный потенциал некоторых тяжелых металлов и микробиологических показателей из фильтратных вод Тбилисского полигона твердых бытовых отходов. Согласно расчетам, сорбционный потенциал загрязняющих веществ из сточных вод при использовании нового сорбента составляет в среднем 60-98%.
Ключевые слова: углеродные материалы, сорбент, термохимическая обработка, фильтрат, очистка, тяжелые металлы, микробиологические показатели.