Desalination and Water Treatment
www.deswater.com

doi: 10.5004/dwt.2023.29408

288 (2023) 234–246
March

Comparative analysis of sorbents within the landfill leachate pretreatment process

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Received 4 October 2022; Accepted 17 February 2023

ABSTRACT

A proper management of leachates, including their treatment and pretreatment, constitutes a challenge for the operators of landfills. A known method of the treatment of leachates is the adsorption. Responding to the need for reasonable management of raw materials and energy, waste is used as substitutes of commercial sorbents. The laboratory tests on the pretreatment of leachates from the landfill in Janczyce included the following compounds: zeolite (Z), activated carbon (AC), shungite (S), walnut shells (WS), spent coffee grounds (SCG) and sewage sludge (SS). The research allowed to determine the influence of the contact time (10, 20, 40, 80 and 160 min) on the chemical oxygen demand (COD), the colour, the dissolved organic compounds and heavy metals at adsorption dosage (5, 10, 20, 40 and 80 g·L^{-1}). The kinetics and isotherms of the adsorption were determined. Better effectiveness of the activated carbon was discovered for the removal of the colour of the leachate and the reduction of the COD in comparison to the shungite and zeolite. Satisfactory effectiveness of sewage sludge, spent coffee grounds and walnut shells was not found for the remediation of landfill leachates. The highest effectiveness of the removal of the COD (13.5% for SS and t = 10 and 20 min; 19.9% for WS and t = 160 min; 12.9% for CG and t = 10 min) and the colour (34.9% for SS and t = 10; 26.6% for WS and t = 20 min) for alternative sorbents was obtained for 5 g·L^{-1} dose. Higher doses and longer time cause the deterioration of the initial parameters of the landfill leachates. More favourable effects of the removal of the COD were obtained for shungite (76% for D = 80 g·L^{-1} and t = 160 min), activated carbon (75.5% for D = 80 g·L^{-1} and t = 10 min) and zeolite (57.5% for D = 40 g·L^{-1} and t = 80 min). In the aspect of the elimination of UVA(254), the greatest results were obtained for activated carbon (99.97%, D = 40 g·L^{-1}, t = 160 min), shungite (74%, D = 80 g·L^{-1}, t = 40 min) and zeolite (14.5%, D = 80 g·L^{-1}, t = 160 min). In the aspect of the elimination of the colour, the greatest results were obtained for activated carbon (99.14%, D = 80 g·L^{-1}, t = 60 min), while slightly weaker effects were obtained for shungite (80.5%, D = 80 g·L^{-1} and t = 80 min) and zeolite (69.25%, D = 80 g·L^{-1}, t = 80 min). An improvement of the adsorption properties of sewage sludge, spent coffee grounds and walnut shells can be realised by proper conditioning.

Keywords: Landfill leachate; Adsorption; Zeolite; Activated carbon; Shungite; Walnut shells; Spent coffee grounds; Sewage sludge; Sorbents

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Presented at the 15th Scientific Conference on Micropollutants in the Human Environment, 14–16 September 2022, Częstochowa, Poland

1. Introduction

In 2021 almost 14 million tones of municipal waste were collected in Poland (a 4.2% increase relative to 2020). The collected waste was sent for recovery (60% – recycling, composting or fermentation, thermal processing) and neutralization through thermal processing without energy recovery (1.3%) and neutralization through landfilling (38.7%). Data indicates that despite the increase of the amount of selectively collected municipal waste (a 25% growth year-to-year), still almost 40% is disposed through storage in landfills [1]. This value does not deviate from the average of 27 European Union member countries, where 38.5% (834 million tones) of waste was sent to landfills. It is estimated that approximately 70% of organic substances reaching landfills is microbiologically transformed and then remains in leachates (10%).

Large leachate volumes are usually generated within 1–2 years from landfilling waste, and after heavy rain [2–4]. In the case of proper landfill management, the volume of generated leachates at the post-operation phase should decrease. Leachates are characterized by a very complex and varied composition, with a high level of pollutants [5]. Chemical parameters are reduced and reach bio-stabilization over time [6]. Fluctuations of such indices as phosphorous, chlorides, calcium, magnesium, sulphate, dissolved solids, heavy metals and BTEX (Benzene, Toluene, Ethylbenzene, Xylenes) depend rather on the season (seasonal variability) than landfill age [7]. Heavy metals, depending on the type, concentration and form, can have a stimulating, neutral or toxic impact on the living organisms [8–17]. Therefore, evaluating the leachate quantity and quality, followed by management and treatment constitute major challenges for all municipal landfill administrators [10] Leachate management can be implemented through [11–13,18–20]: A – transporting leachates and treatment jointly with municipal sewage (most common in Poland due to simplicity and minor costs); B – bio-treatment under aerobic or anaerobic conditions; C – treatment with physical and chemical methods; D – hydrophobic treatment, E – combination of the methods or other. One of the physico-chemical methods applied for the treatment of leachate is adsorption, which enables removing biodegradable substances, humic substances and chlorinated hydrocarbons. Porous substances, for example, activated carbon, due to their large surface area, are particularly advantageous adsorptive materials. The adsorption process continues until achieving the dynamic equilibrium in the concentration of the adsorbate remaining in the solution and found on the internal surface of the adsorbent. The speed of this process depends on, among others, adsorbent particle size – the larger the particles, the slower the adsorption process [14].

Correct utilization of natural resources is currently a priority, and the Framework Directive on waste refers to the "4R" principle or the so-called recovery hierarchy, namely, Reduce, Reuse, Recycle, Recover, which encourages to rationally consume waste and energy [15]. This is the reason why cheaper substitutes for commercial adsorbents are sought among waste. Some of the non-conventional adsorbents are activated carbons produced from bamboo dust, bark husk, chitin, coir pith, lignite, maize cob, palm shell, palm fibre, palm stone, peat, pinewood, rice husk, sago waste, sawdust, sugarcane bagasse and tea leaves. Furthermore, the used adsorbents include zeolites, activated aluminium oxide, ash from municipal waste or sewage sludge incineration, resins, etc. [16,17]. A considerable advantage of non-conventional adsorbents is that they are renewable and more eco-friendly compared to conventional activated carbons. Moreover, they are able to achieve almost a similar treatment efficiency [21,22]. The advantages of the solid body adsorption process include low costs, process simplicity and no by-products [17]. However, in the case of activated carbon adsorption, ensuring high process effectiveness with simultaneous minimizing of regeneration time and costs requires pretreatment of leachates [23]. A significant disadvantage of activated carbon is its high cost, which is the reason why inexpensive, readily available adsorbents are sought after [22].

The aim of the research was to analyse the effectiveness of the pretreatment of landfill leachate with the use of commercial sorbents and their alternative substitutes. So far, there have been no tests on the pretreatment of leachates from the landfill in Janczyce with the use of sorbents, including waste sorbents. The landfill in Janczyce is representative in terms of its technical-operational properties with regard to the management of municipal sewage and landfill leachates. The experience gained from the treatment of leachates from this particular landfill can be applied in the treatment of leachates from other operating municipal waste landfills of similar characteristics. A significant element of the research is also the utilization of wastes as sorbents in accordance with the "4R" principal (Reduce, Reuse, Recycle, Recover), which encourages to rationally consume waste and energy.

2. Material and methods

2.1. Landfill leachate and sorbents

Leachate was collected at the landfill in Janczyce (the town located in the south–eastern part of Poland). The landfill is part of the Municipal Waste Treatment Plant in Janczyce and has been operating since 2003. The facility handles approximately 150,000 residents, and almost 300 kg waste per capita per year is generated within the region. After the mechanical treatment of mixed communal waste, sorting residues and other waste unsuitable for the recovery, waste is stored within the landfill area (3.64 ha). More than 6,600 m^3 of leachate is generated on average per year in association with landfill operation.

Laboratory testing of landfill leachate pretreatment was conducted with the use of six sorbents: zeolite (Z), activated carbon (AC), shungite (S), walnut shells (WS), spent coffee grounds (SCG) and sewage sludge (SS) (Fig. 1, Table 1.)

2.2. Sampling preparation

Laboratory tests involving landfill leachate pretreatment were conducted on leachate samples with a volume of 0.1 L (sample volume selected, so that it was possible to analyse all determined parameters; Fig. 2). The following sorbent doses were weighed, respectively to 0.2 L Erlenmeyer flasks – L^{-1}, 10, 20 and 40, 80 $g{\cdot}L^{-1}$. These were then supplemented with 0.1 L of leachates, followed by mixing in a

shaker at the speed of 100 rpm^{-1} and over a specified period of time. Contact times of 10, 20, 40, 60, 80 and 160 min were applied. Afterwards, the samples were filtered and tested towards the presence of the following: chemical oxygen demand (COD), colour, dissolved organic compounds and heavy metals (Fig. 2).

2.3. Analytical methods

Collecting leachate samples, their storage and tests on the chemical composition were conducted in accordance with the applicable PN-ISO 5667-10:2021-11 standard.

The pH value of the sorbents was determined with the potentiometric method in a suspension of sorbent and electrolyte (potassium chloride in a concentration of 1 mol·L^{-1}; pH_{KCl}) with the use of a Mettler Toledo pH meter, in accordance with PN-ISO 10390:1997. The contribution of the following chemical elements C, H, N, S, O of sorbents was determined using a CHNS/O FlashSmart-series elemental analyser (Thermo Fisher Scientific, Waltham, USA). The particle size distribution of sorbents was analysed with the use of a Mastersizer 3000 laser particle size analyser by Malver, which measures particle size in the range from 0.01 to 3,500 μm. Sorbents were also analysed with the FEI QUANTA 250 FEG (FEI, Brno, Czech Republic) scanning microscope (Fig. 5). The surface area of sorbents was calculated through the Multi Brunauer–Emmett–Teller (multi-point

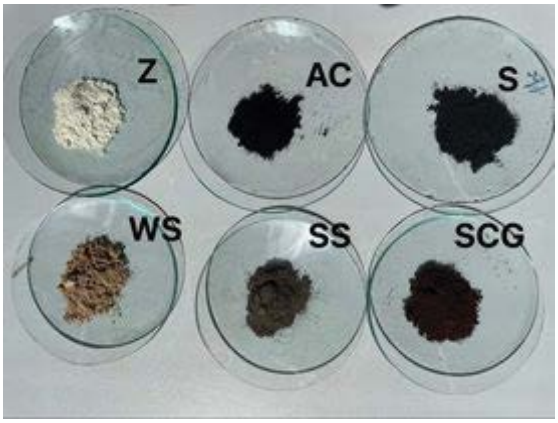

Fig. 1. Sorbents used within the landfill leachate pretreatment process Z – zeolite, AC – activated carbon, S – shungite, WS – walnut shells, SS – sewage sludge, SCG – spent coffee grounds.

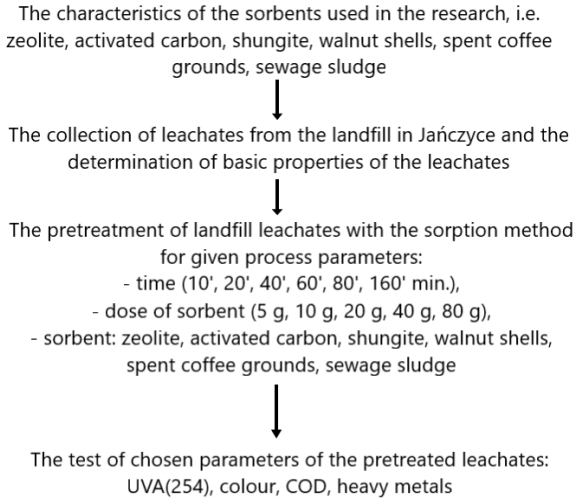

Fig. 2. Research procedure.

Table 1
Sorbents used

Activated carbon (AC)	Commercial NORIT SX 2 p.a. grade powder activated carbon by CHEMPUR Company, (Piekary Śląskie, Poland). *substances dissolved in HCl – max. 1.0%, LOI max. (110°C) 10%, Cu max. 0.001%, Ca max. 0.02%, Fe max. 0.02%, molasses number max. 380. Particle size (100%) < 399.55 μm (Figs. 3 and 4)
Zeolite (Z)	Commercial product by NatVita, white-cream colour. Composition*: 100% zeolite, including clinoptilolite >84%, cristobalite 8%, clay minerals 4%, plagioclase 3%–4%, rutile 0.1%–0.3%. Particle size (100%) < 515.77 μm (Figs. 3 and 4)
Shungite (S)	Commercial product; sedimentary rock of organic origin. Composition*: silicon oxide (approx. 45%), carbon (approx. 30%), silicates (approx. 5%), sulfides (approx. 20%), as well as trace amounts of fullerenes (0.0001%–0.001%). Commercial diameter of 10–20 mm. Shungite was dried at the temperature of 105°C, ground in a ball mill and pounded in a mortar. The above-mentioned allowed to obtain a sorbent with a particle size (100%) < 40.15 μm (Figs. 3 and 4)
Walnut shells (lat. *Juglans regia*) (WS)	Shells of walnut commonly growing in Poland. Shells were dried to dry mass at a temperature of 105°C, ground in a ball mill and pounded in a mortar. This enabled achieving a sorbent with a particle size (100%) of <859.44 μm (Figs. 3 and 4)
Spent coffee grounds (SCG)	Obtained after brewing coffee. Grounds were dried to dry mass at 105°C, and then pounded in a mortar. This enabled achieving a sorbent with a particle size (100%) < 756.45 μm (Figs. 3 and 4)
Sewage sludge (SS)	Stabilized, drained and dried in a disc dryer, collected from a mechanical and biological wastewater treatment plant at Sitkówka-Nowiny (PE = 289,000; $Q_{dśr}$ = 51,000 m^3·d^{-1}). Sewage sludge was dried at a temperature of 105°C and pound in a mortar. This enabled achieving a sorbent with a particle size (100%) < 211.07 μm (Figs. 3 and 4)

*According to manufacturer's information.

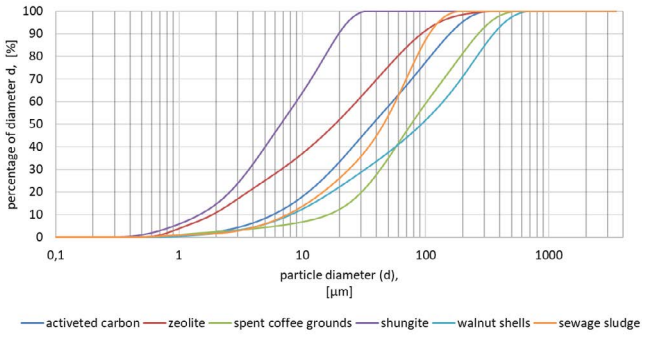

Fig. 3. Contribution of sorbents with a diameter of *d* (%).

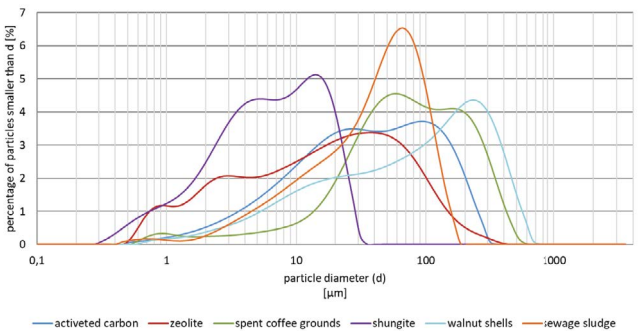

Fig. 4. Contribution of sorbents with a diameter smaller than *d* (%).

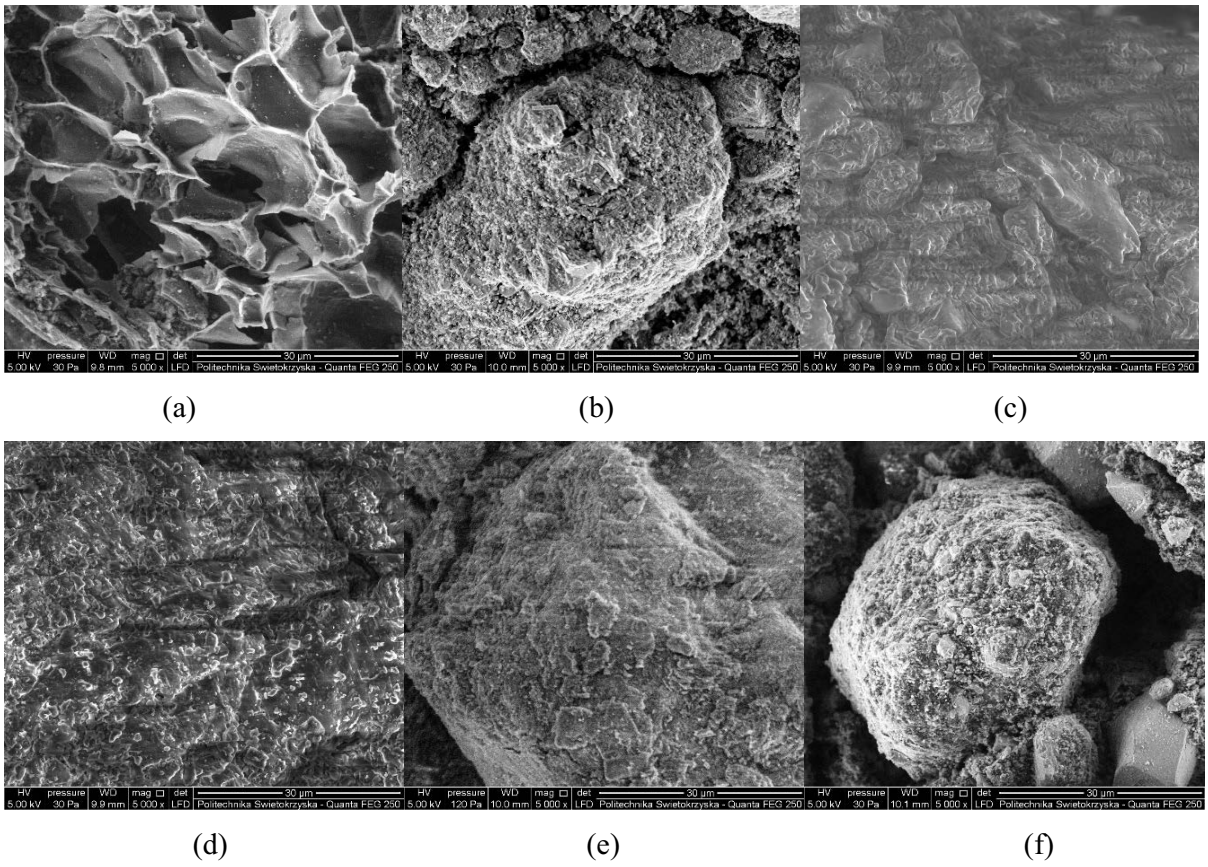

Fig. 5. Scanning electron microscopy of used sorbents: (a) activated carbon, (b) zeolite, (c) spent coffee grounds, (d) walnut shells, (e) sewage sludge and (f) shungite.

BET) method and using AutosorbiQ-MP-MP (2 Stat.; Viton by Quantachrome Instruments (Anton Paar) from Boynton Beach, Florida, USA). Loss on ignition (LOI) of sorbents was determined in accordance with PN-EN 15935:2022-1, at the temperature of 550°C. The phase composition of used sorbents was determined with the Debye–Scherrer–Hull X-ray powder diffraction (Fig. 6). Empyrean diffractometer was used for this purpose (PANalytical, Almeo, Niederlande). The test was conducted in the range of angles 6°–70° 2θ with the use of a Cu lamp and the X'Celerator strip detector. The interpretation of the test results was performed in the High-Score Plus programme (PANalytical, Almeo, Niederlande).

Heavy metal content in the prepared samples was determined using the ICP-OES Optima 8000 spectrophotometer (PerkinElmer, Waltham, USA), in accordance with ISO 9001: 2000. Aqua regia mineralization was conducted in compliance with the PN-EN ISO 15587-1:2005 standard, with the following parameters applied:

- a 25 cm^3 leachate sample was supplemented with: 7.5 cm^3 HCl p.a. grade (1.19 $g \cdot cm^{-3}$, 38%) and 2.5 cm^3 HNO_3 p.a. grade (1.40 $g \cdot cm^{-3}$, 65%),
- the sample was heated for approx. 30 min under a watch glass,

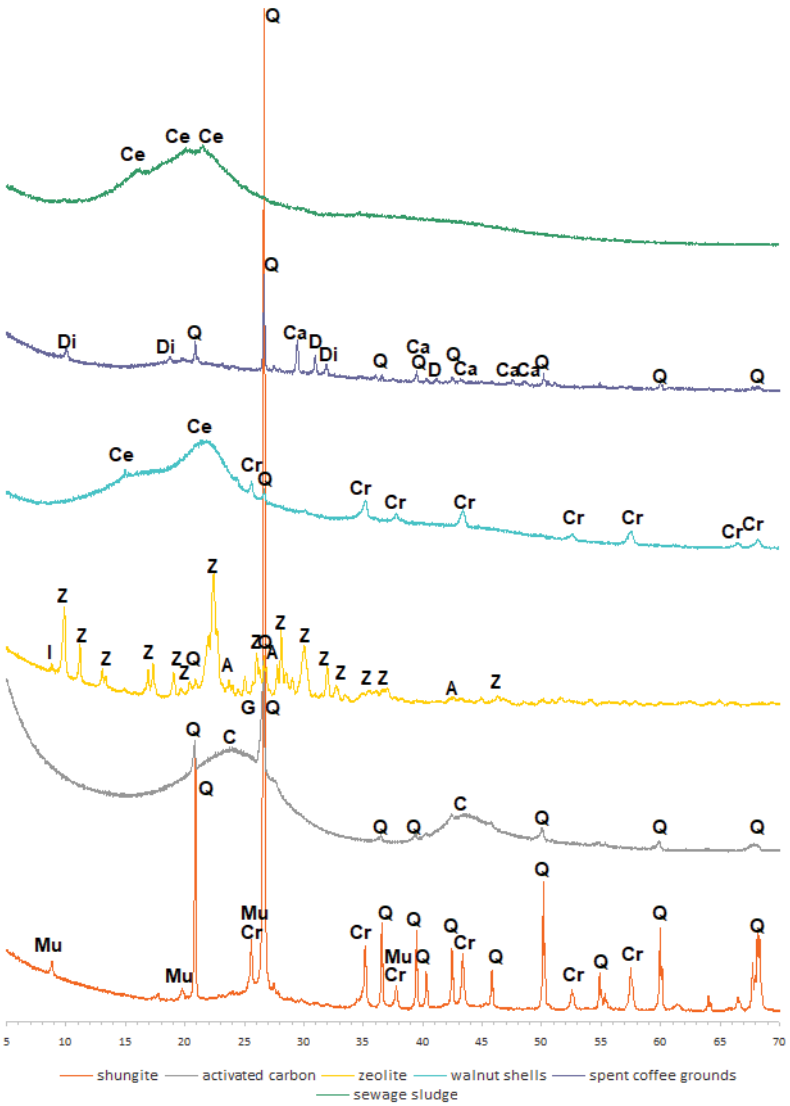

Fig. 6. X-ray patterns of shungite, activated carbon, zeolite, walnut shells, spent coffee grounds and sewage sludge; A - anortic (plagioclase), C - carbon, Ca - calcite, Ce - cellulose, Cr - corundum, D - dolomite, Di - dittmarite, G - graphite, I - illite, Mu - muscovite, Q - quartz, Z - clinoptilolite.

- next, the contents was evaporated until it was almost dry, the residue was flooded with 20 cm^3 p.a. grade HNO_3 with a concentration of 5%,
- the cooled solution was quantitatively transferred to a 50 cm^3 volumetric flask and supplemented with 50 cm^3 of redistilled water.

Chemical oxygen demand of leachates was determined with the Spectroquant photometric method – determination through cuvette testing using a Spectroquant Nova 60 spectrophotometer (MERCK, Darmstadt, Germany) after prior heating in a TR 320 thermo-reactor by MERCK (120 min at 148°C). The determination limit for the selected method was 300 to 3,500 $mg \cdot L^{-1}$ (Standard Deviation of Procedure ±13.9 $mg \cdot L^{-1}$).

The colour was determined with the spectrophotometric method using a UV/VIS spectrophotometer, in accordance with PN-EN ISO 7887:2012, while dissolved organic compounds were determined through UVA(254) spectrophotometry (wavelength – 254 nm) as per the PN-C 04572:1984 standard. Both parameters were tested using the Genesis 150 UV-VIS spectrophotometer (Thermo Fisher Scientific, Waltham, USA).

The pH value was determined with a potentiometric method using a CX-505 Multifunction Meter (METTLER TOLEDO, Columbus, USA), pursuant to PN-EN ISO 10523:2012.

3. Results and discussion

Leachate sample was: pH 7.975, COD 1,742 $mgO_2 \cdot L^{-1}$, colour 2,442 $mgPt \cdot L^{-1}$, UVA(254) 3.435.

Landfill leachate test results with respect to the COD are shown in Figs. 7–11, while the variability of UVA(254) and colour indices are shown accordingly in Table 3. Due to the extensive structure of the experiment, the authors decided

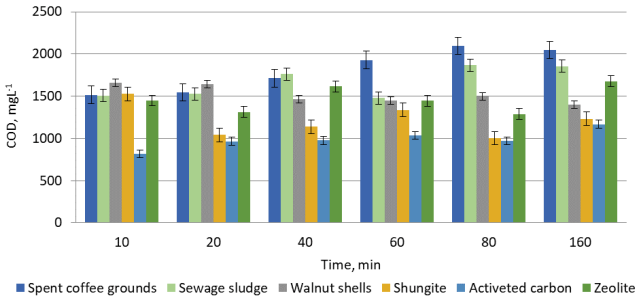

Fig. 7. Impact of the adsorption duration on the chemical oxygen demand of landfill leachates at an arbitrarily adopted adsorbent dose of $D = 5\ g \cdot L^{-1}$.

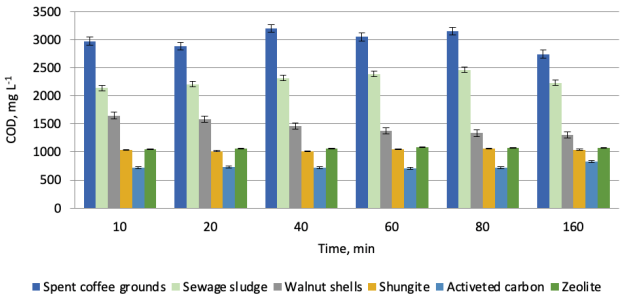

Fig. 8. Impact of the adsorption duration on the chemical oxygen demand of landfill leachates at an arbitrarily adopted adsorbent dose of $D = 10\ g \cdot L^{-1}$.

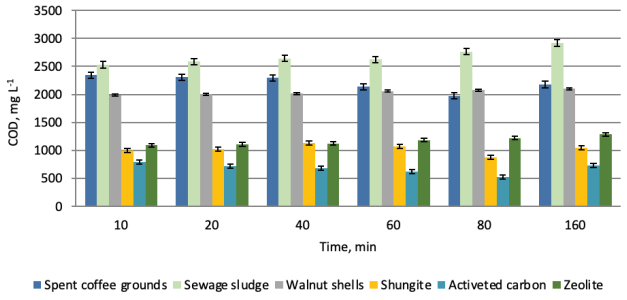

Fig. 9. Impact of the adsorption duration on the chemical oxygen demand of landfill leachates at an arbitrarily adopted adsorbent dose of $D = 20\ g \cdot L^{-1}$.

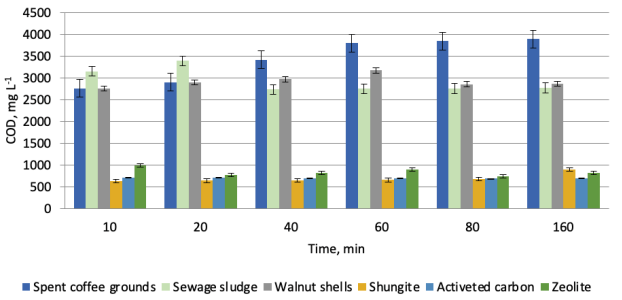

Fig. 10. Impact of the adsorption duration on the chemical oxygen demand of landfill leachates at an arbitrarily adopted adsorbent dose of $D = 40\ g \cdot L^{-1}$.

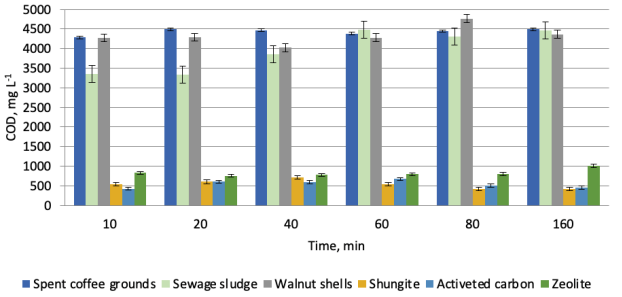

Fig. 11. Impact of the adsorption duration on the chemical oxygen demand of landfill leachates at an arbitrarily adopted adsorbent dose of $D = 80\ g \cdot L^{-1}$.

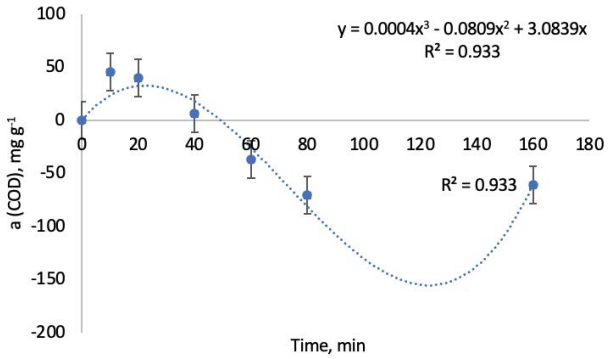

Fig. 12. Adsorption kinetics of coffee grounds at an arbitrarily adopted adsorbent dose $D = 5\ g \cdot L^{-1}$.

to only show selected adsorption isotherms for systems where the highest COD, UVA(254) and colour elimination degree were achieved.

The adsorptive properties of dried coffee grounds as an adsorbent seem negligible. This is demonstrated in Fig. 12, where a certain improvement in leachate quality was obtained at a minor dose $D = 5\ g \cdot L^{-1}$ and contact times of 10, 20 and 40 min. In the remaining cases, effluent composition significantly deteriorated, while an increase in colour and UVA(254) was recorded in each case (Table 4), which is marked in bold text (Table 3). Much more favourable effects of the removal of the COD and colour (>90%; maximal adsorption capability 40 mm of the COD per 1 g of activated carbon from coffee grounds) were obtained by Ferraz and Yuan [24] by using activated carbon from coffee grounds saturated with H_3PO_4 – the surface 188–2,118 $m^2 \cdot g^{-1}$. Also Carvajal-Florez and Oakley [25] could see a great potential for sewage treatment in coffee pulp, however it is necessary to apply such sorbent of chemical and thermal modification in order to improve the physico-chemical state and the sorption capability

Sorptive properties of coffee grounds could be somewhat improved, provided that they undergo a tedious process of conditioning and/or rinsing with distilled water.

A similarly restricted adsorption effectiveness was achieved when using sewage sludge as an adsorbent. Also in

Table 2
Characteristics of used sorbents

Sorbent	pH (–)	Colour (–)	Odour (–)	LOI (%)	BET surface area ($m^2 \cdot g^{-1}$)	C (%)	H (%)	N (%)	S (%)	O (%)
SCG	4.70	Dark brown	Coffee	96.90	48.55	50.14 ± 0.19	6.63 ± 0.08	2.78 ± 0.01	0.10 ± 0.01	36.84 ± 0.62
SS	6.29	Grey	Specific, unpleasant	67.36	31.18	34.37 ± 0.18	5.04 ± 0.06	4.97 ± 0.01	1.17 ± 0.01	25.26 ± 0.61
WS	3.89	Light brown	Slightly nutty	90.37	51.44	44.44 ± 0.43	4.40 ± 0.19	0.44 ± 0.02	0.0 ± 0.0	38.01 ± 0.95
S	5.05	Black	Odourless	25.16	46.92	24.29 ± 0.16	0.24 ± 0.003	0.20 ± 0.003	0.26 ± 0.01	1.78 ± 0.03
AC	8.92	Black	Odourless	93.71	677.53	88.26 ± 1.24	0.47 ± 0.02	0.38 ± 0.04	0.08 ± 0.07	3.01 ± 0.36
Z	6.59	White, slightly creamy	Odourless	5.42	50.35	0.03 ± 0.01	0.99 ± 0.17	0.0 ± 0.0	0.001 ± 0.002	7.81 ± 0.86

this case, it is easily noticeable that leachate quality tends to deteriorate with longer exposure times and increased dose. This is demonstrated in Fig. 13, where a certain improvement in leachate quality was obtained only at a low dose D = 5 $g \cdot L^{-1}$ and contact times of 10 and 20 min (approx. 13.5% of the COD). In terms of decolourization, remediation results were much better, since improved leachate quality was achieved regardless of the process duration, and the optimal results were obtained for time t = 10 min (Fig. 13) and the authors believe the recorded colour reduction was significant, and amounted to 34.9%. In other cases, when the dose was D > 5 $g \cdot L^{-1}$, the effluent COD was clearly increased, which was shown in Figs. 8–11. Greater adsorbent dose entailed increased leachate UVA(254) and colour. Similar results in this respect were recorded for walnut shells. In this case, and for time t = 160 min (Fig. 14), it was possible to reduce the COD proportionally to contact time, down to a value of approximately 19.9%. A slightly better effect (26.6%) was achieved with respect to decolourization. However, extended exposure times entailed deteriorated decolourization results, even at t = 20 min (Table 3). Higher adsorbent dose led to a significant reduction in leachate quality indices (Table 4). Latosińska et al. [26] takes notice of the fact, that the introduction of the alternative sorbents (e.g., sewage sludge ash) into the environment without prior pretreatment causes a great risk of the accumulation of metals. According to the conducted research – high doses of sewage sludge and longer time of exposition can deteriorate the state of the pre-treated leachates. It is worth to consider in further tests to conduct both the physical (carbon dioxide, steam) and chemical activations (KOH, NaOH, $ZnCl_2$, etc.) in order to increase the porosity and the surface of sewage sludge [27]. The use of $KMnO_4$ for the modification of the activated carbon increases its surface area and the volume of pores more efficiently in comparison to the modification of HNO_3 and heating [28].

In terms of eliminating the COD, the highest efficiency was obtained for a sample where landfill leachates were exposed to a shungite dose D = 80 $g \cdot L^{-1}$ for 160 min (Fig. 11). The result was the elimination of 76% of the COD, down to a value of 418 $mg \cdot L^{-1}$ COD. However, for an exposure time equal to 80 min, the recorded result was comparable – the COD at a level of 422 $mg \cdot L^{-1}$. High efficiency in this respect was also obtained in the case of activated carbon, where for a dose D = 80 $g \cdot L^{-1}$ and only 10 min of adsorption, the recorded COD equalled 426 $mg \cdot L^{-1}$ and a 75.5% process efficiency (Fig. 11). According to Kulikowska and Sułek [29] the process of adsorption with the use of powdery activated carbon (PAC) Norit SX2 is also more effective within the first 10 min of the process. The effectiveness of the removal of the COD varied within the range from 30.9% to 83.3% with the PAC dose from 2 to 10 $g \cdot L^{-1}$ and the initial value of the COD 1,007 $mg \cdot L^{-1}$ [29]. The research by Abuabdou et al. [30] also proved high effectiveness of the removal of the COD and colour (85.47% and 95.65% respectively) from landfill leachates (the COD 1,452.5 $mg \cdot L^{-1}$, pH value 8.89, colour 1525 $mg \cdot Pt \cdot L^{-1}$, temperature 28.45°C) with the use of the adsorption on the activated carbon under optimal conditions (the speed of shaking 250 rpm^{-1}, the contact time of 4h, dose 40 $g \cdot L^{-1}$). A significant disadvantage of the activated carbon is its cost. Therefore, less costly and easily

Table 3
Test results with respect to the chemical oxygen demand, UVA(254) and colour in leachates after the adsorption process using coffee grounds, sewage sludge and walnut shells $D = 5\ g \cdot L^{-1}$

Coffee grounds			
Time, (min)	Chemical oxygen demand, ($mg \cdot L^{-1}$)	UVA(254)	Colour, ($mg \cdot L^{-1}$)
0	1,742	3.435	2,442
10	1,516	***3.865***	***2,448***
20	1,544	***3.830***	***2,639***
40	1,712	***4.006***	***2,809***
60	***1,928***	***4.026***	***2,957***
80	***2,096***	***4.055***	***2,972***
160	***2,048***	***3.908***	***2,938***
Sewage sludge			
Time, (min)	Chemical oxygen demand, ($mg \cdot L^{-1}$)	UVA(254)	Colour, ($mg \cdot L^{-1}$)
0	1,742	3.435	2,442
10	1,506	***3.581***	1,589
20	1,526	***3.657***	1,782
40	***1,760***	***3.586***	1,893
60	1,478	***3.647***	1,896
80	1,866	***3.665***	1,955
160	1,854	***3.687***	1,995
Walnut shells			
Time, (min)	Chemical oxygen demand, ($mg \cdot L^{-1}$)	UVA(254)	Colour, ($mg \cdot L^{-1}$)
0	1,742	3.435	2,442
10	1,660	***3.750***	1,793
20	1,640	***3.764***	1,881
40	1,464	***3.758***	1,853
60	1,450	***3.756***	1,807
80	1,496	***3.841***	1,933
160	1,396	***3.806***	2,114

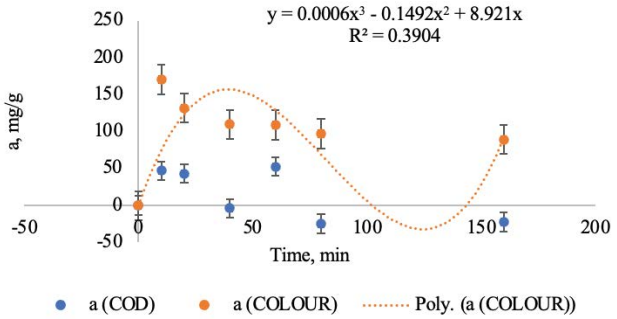

Fig. 13. Adsorption kinetics of sewage sludge at an arbitrarily adopted adsorbent dose $D = 5\ g \cdot L^{-1}$.

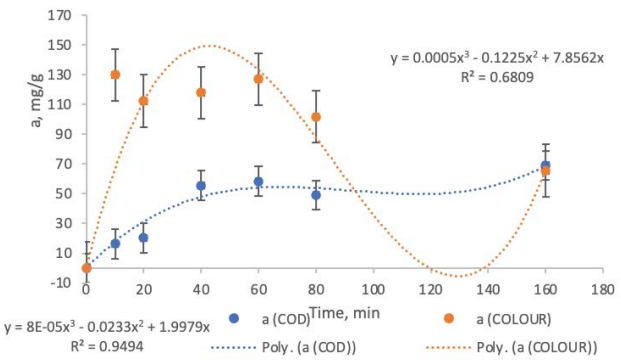

Fig. 14. Adsorption kinetics of walnut shells at an arbitrarily adopted adsorbent dose $D = 5\ g \cdot L^{-1}$.

accessible adsorbents are sought for [22]. With such a short contact time, shungite achieved efficiency below 68.8%. The highest efficiency obtained for a zeolite sorbent was the COD elimination index of 57.5%. There was 740 $mg \cdot L^{-1}$ COD left in leachates after an 80-min process, at a dose $D = 40\ g \cdot L^{-1}$ (Fig. 10). As far as UVA(254) elimination is concerned, the highest efficiency was recorded for a sample, where leachates were exposed to a $D = 40\ g \cdot L^{-1}$ dose of activated carbon for 160 min. As an outcome, 99.97% of UVA(254) was removed from the leachates, with the value of this index equal to 0.001 (Table 4). Increasing the activated carbon dose to 80 $g \cdot L^{-1}$ did not alter the remediation effects, while reducing it to

Table 4
UVA(254) and colour test results for landfill leachate samples with an exposure time of 10–160 min and an adsorbent dose of 5–80 g·L^{-1}

Time	UVA(254)	Colour	Time	UVA(254)	Colour	Time	UVA(254)	Colour	Time	UVA(254)	Colour	Time	UVA(254)	Colour
min	–	mg·L^{-1}	min	–	mg·L^{-1}	min	–	mg·L^{-1}	min	–	mg·L^{-1}	min	–	mg·L^{-1}
Coffee grounds														
Dose 5 g			Dose 10 g			Dose 20 g			Dose 40 g			Dose 80 g		
10′	3.865	2,448	10′	4.263	4,312	10′	5.000	6,549	10′	5.000	7,198	10′	5.000	8,000
20′	3.83	2,639	20′	4.660	4,389	20′	5.000	6,599	20′	5.000	8,000	20′	5.000	8,000
40′	4.006	2,809	40′	4.424	4,583	40′	4.545	6,471	40′	5.000	8,000	40′	5.000	8,000
60′	4.026	2,957	60′	4.220	4,894	60′	4.964	6,952	60′	5.000	8,000	60′	5.000	8,000
80′	4.055	2,972	80′	4.568	4,901	80′	5.000	7,972	80′	5.000	8,000	80′	5.000	8,000
160′	3.908	2,938	160′	4.908	5,863	160′	5.000	7,867	160′	5.000	8,000	160′	5.000	8,000
Sewage sludge														
Dose 5 g			Dose 10 g			Dose 20 g			Dose 40 g			Dose 80 g		
10′	3.581	1,589	10′	3,697	2,062	10′	3.855	2,417	10′	3.988	2,823	10′	4.315	3,566
20′	3.657	1,782	20′	3.750	1,999	20′	3.881	2,934	20′	4.013	3,158	20′	4.253	3,369
40′	3.586	1,893	40′	3.769	1,796	40′	4.072	3,157	40′	4.057	3,657	40′	4.002	3,116
60′	3.647	1,896	60′	3.753	2,069	60′	3.837	2,572	60′	4.143	3,856	60′	3.999	3,322
80′	3.665	1,955	80′	3.790	2,588	80′	3.905	2,931	80′	4.111	3,319	80′	4.012	3,477
160′	3.687	1,995	160′	3.700	2,295	160′	4.043	3,227	160′	4.133	3,395	160′	3.995	3,555
Walnut shells														
Dose 5 g			Dose 10 g			Dose 20 g			Dose 40 g			Dose 80 g		
10′	3.75	1,793	10′	3.953	2,231	10′	4.004	2,707	10′	4.341	3,480	10′	4.298	3,608
20′	3.764	1,881	20′	3.930	2,454	20′	4.042	2,687	20′	4.229	3,206	20′	4.376	3,892
40′	3.758	1,853	40′	3.903	2,958	40′	4.224	2,735	40′	4.355	3,156	40′	4.293	3,666
60′	3.756	1,807	60′	3.938	2,417	60′	3.961	2,872	60′	4.047	3,923	60′	4.322	3,700
80′	3.841	1,933	80′	4.159	2,847	80′	4.101	3,053	80′	4.369	3,666	80′	4.375	3,661
160′	3.806	2,114	160′	3.977	2,487	160′	4.114	3,489	160′	4.263	3,626	160′	4.500	4,125
Shungite														
Dose 5 g			Dose 10 g			Dose 20 g			Dose 40 g			Dose 80 g		
10′	3.291	1,742	10′	2.733	901	10′	2.297	831	10′	1.503	713	10′	0.803	493
20′	3.235	1,763	20′	2.732	961	20′	2.046	643	20′	1.551	724	20′	1.183	585
40′	3.314	1,767	40′	2.649	976	40′	1.876	691	40′	1.693	704	40′	0.893	499
60′	3.356	1,749	60′	2.723	853	60′	2.039	681	60′	1.641	699	60′	0.984	485
80′	3.371	1,786	80′	2.707	998	80′	3.951	908	80′	1.617	647	80′	1.106	476
160′	3.334	1,760	160′	2.790	956	160′	2.325	923	160′	1.601	705	160′	1.051	537
Activated carbon														
Dose 5 g			Dose 10 g			Dose 20 g			Dose 40 g			Dose 80 g		
10′	1.042	625	10′	0.617	95	10′	0.294	71	10′	0.109	49	10′	0.031	43
20′	1.024	477	20′	0.522	84	20′	0.234	102	20′	0.022	37	20′	0.062	42
40′	1.263	673	40′	0.547	86	40′	0.274	125	40′	0.030	27	40′	0.012	23
60′	1.133	677	60′	0.530	82	60′	0.195	118	60′	0.018	32	60′	0.001	21
80′	1.162	547	80′	0.565	80	80′	0.181	116	80′	0.003	39	80′	0.008	33
160′	1.510	641	160′	0.422	79	160′	0.100	106	160′	0.001	33	160′	0.001	41
Zeolite														
Dose 5 g			Dose 10 g			Dose 20 g			Dose 40 g			Dose 80 g		
10′	3.414	1,668	10′	3.338	1,056	10′	3.261	961	10′	3.186	983	10′	3.071	881
20′	3.409	1,645	20′	3.325	1,106	20′	3.187	1,008	20′	3.284	1,053	20′	3.067	887
40′	3.416	1,771	40′	3.291	1,062	40′	3.274	1,007	40′	3.251	1,056	40′	3.051	822
60′	3.427	1,708	60′	3.322	1,094	60′	3.203	927	60′	3.249	1,023	60′	3.057	809
80′	3.447	1,650	80′	3.376	1,068	80′	3.236	927	80′	3.240	1,008	80′	2.947	751
160′	3.459	1,668	160′	3.201	954	160′	3.075	987	160′	3.032	879	160′	2.936	798
Raw landfill leachate				3.435	2,442									

Table 5
Test results for heavy metal tests in leachate samples, for an adsorbent dose D = 80 g·L^{-1} and a variable exposure time t = 10, 20, 40, 80 and 160 min

Time	Cd	Cu	Cr	Ni	Pb	Zn	Co
min	mg·L^{-1}	mg·L^{-1}	mg·L^{-1}	mg·L^{-1}	mg·L^{-1}	mg·L^{-1}	mg·L^{-1}
Spent coffee grounds							
10′	0.0001	0.2609	0.0761	0.1001	0.0516	0.4875	0.1367
20′	0.0101	0.2718	0.0727	0.1028	0.1582	0.4818	0.1546
40′	0.0005	0.2844	0.0737	0.1053	0.0404	0.4545	0.1500
60′	0.0110	0.2919	0.0821	0.1184	0.2026	0.6114	0.1595
80′	0.0001	0.3910	0.0815	0.1139	0.0670	0.4713	0.1629
160′	0.0064	0.3829	0.0759	0.1148	0.1968	0.5239	0.1793
Sewage sludge							
10′	0.0274	0.5140	0.0764	0.2679	0.3005	0.7436	0.0580
20′	0.0001	0.5986	0.0670	0.1617	0.0289	0.4765	0.0290
40′	0.0001	0.5628	0.0682	0.1625	0.0300	0.4026	0.0304
60′	0.0001	0.9508	0.0724	0.2877	0.0363	0.5936	0.0537
80′	0.0001	0.7769	0.0937	0.2382	0.0286	0.6928	0.0499
160′	0.0001	0.8943	0.0675	0.2714	0.0321	0.6387	0.0574
Walnut shells							
10′	0.0001	0.3592	0.0665	0.0916	0.0303	0.4818	0.0053
20′	0.0001	0.1265	0.0936	0.0788	0.0172	0.4104	0.0068
40′	0.0008	0.2014	0.0655	0.0685	0.0316	0.4613	0.0030
60′	0.0001	0.1982	0.0648	0.0660	0.0566	0.3409	0.0025
80′	0.0001	0.2035	0.0660	0.0682	0.0252	0.4015	0.0049
160′	0.0055	0.1976	0.0598	0.0721	0.1061	0.4632	0.0120
Shungite							
10′	0.0008	0.0617	0.0132	0.0636	0.0488	0.2144	0.0001
20′	0.0001	0.0319	0.0156	0.0613	0.0245	0.2085	0.0001
40′	0.0016	0.0539	0.0148	0.0682	0.0441	0.2095	0.0007
60′	0.0001	0.0377	0.0423	0.0606	0.0222	0.2640	0.0036
80′	0.0001	0.0432	0.0149	0.0643	0.0216	0.2205	0.0001
160′	0.0321	0.0518	0.0126	0.0595	0.3644	0.2729	0.0080
Activated carbon							
10′	0.0125	0.0195	0.0061	0.0209	0.1151	0.1644	0.0001
20′	0.0088	0.0663	0.0119	0.0036	0.0386	0.2143	0.0001
40′	0.0059	0.0287	0.0058	0.0121	0.0908	0.1451	0.0001
60′	0.0001	0.0296	0.0082	0.0122	0.0143	0.2131	0.0001
80′	0,0001	0.0193	0.0057	0.0135	0.0109	0.1274	0.0001
160′	0.0001	0.0173	0.0051	0.0121	0.0264	0.1446	0.0006
Zeolite							
10′	0.0005	0.0346	0.0516	0.0800	0.0434	0.8959	0.0038
20′	0.0011	0.0249	0.0758	0.0683	0,0377	0.3080	0.0037
40′	0.0274	0.0286	0.0507	0.0651	0.2671	0.2497	0.0050
60′	0.0008	0.0287	0.0489	0.0603	0.0386	0.2482	0.0007
80′	0.0001	0.0665	0.0558	0.0825	0.0311	0.3718	0.0029
160′	0.0032	0.0379	0.0455	0.0745	0.0926	0.6397	0.0039
Raw landfill leachate	0.0087	0.1269	0.0780	0.0549	0.0607	0.3006	0.0062

20 g·L^{-1} led to a 100-fold increase in UVA(254). The highest efficiency obtained for shungite is a UVA(254) elimination index equal to 74.0%, obtained for an exposure time equal to 40 min and a dose D = 80 g·L^{-1}. In the case of zeolite, the UVA(254) elimination result is as follows – the highest efficiency (14.5%) was obtained for D = 80 g·L^{-1} and an exposure time of 160 min. In this respect, zeolite effectiveness is significantly behind the two sorbents above, however, it is clearly superior to sewage sludge, coffee grounds and walnut shells.

In terms of eliminating leachate colour, the highest efficiency was obtained with an 80 g·L^{-1} dose of activated carbon and an exposure of 60 min. This provided a colour

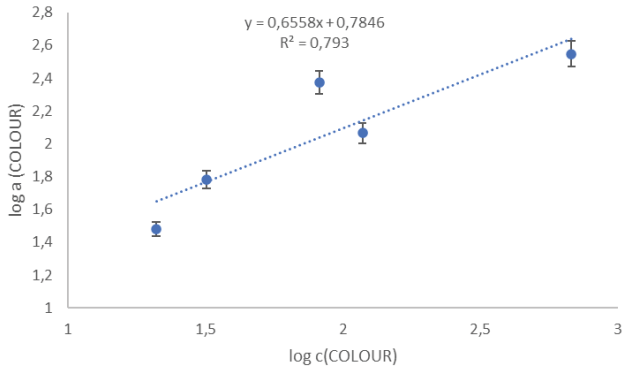

Fig. 15. Relationship between the logarithm of adsorbent concentration on activated carbon surface and in the solution, for leachate colour after t = 60 min.

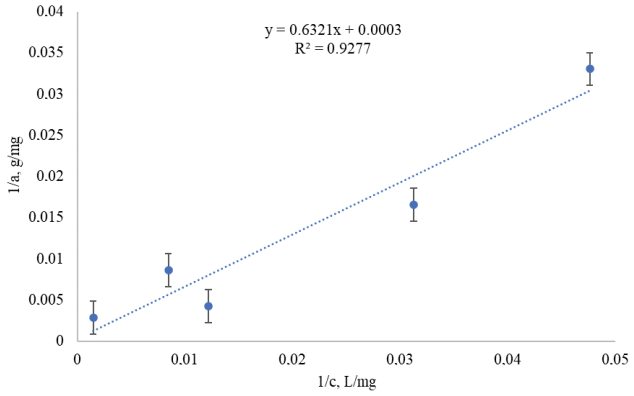

Fig. 16. Relationship between the converse of adsorbent concentration on activated carbon surface and in the solution, for leachate colour after t = 60 min.

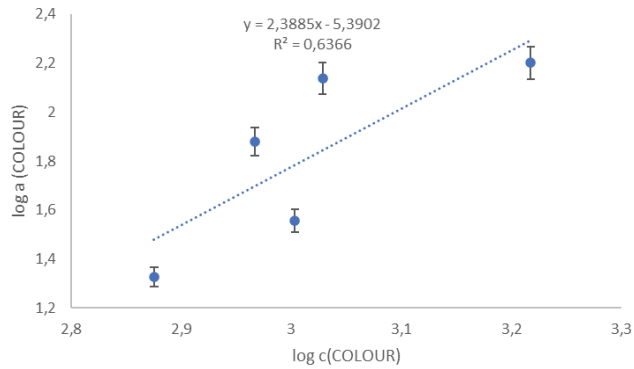

Fig. 17. Relationship between the logarithm of adsorbent concentration on zeolite surface and in the solution, for leachate colour after t = 80 min.

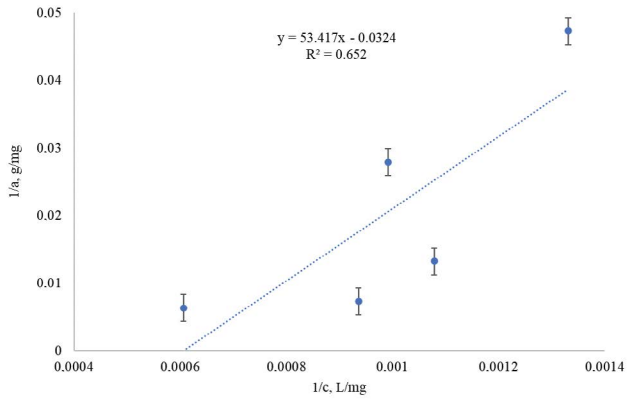

Fig. 18. Relationship between the converse of adsorbent concentration on zeolite surface and in the solution, for a leachate colour after t = 80 min.

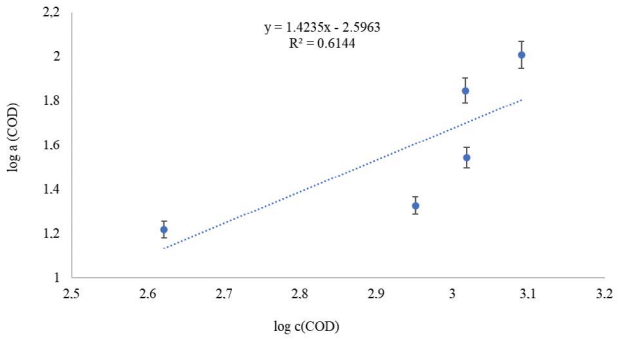

Fig. 19. Relationship between the logarithm of adsorbent concentration on shungite surface and in the solution, for leachate chemical oxygen demand after t = 160 min.

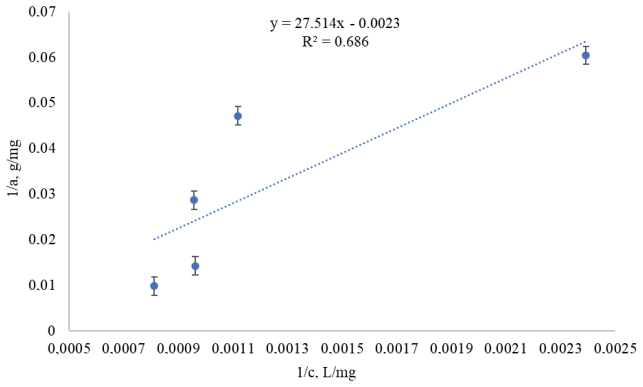

Fig. 20. Relationship between the converse of adsorbent concentration on shungite surface and in the solution, for leachate chemical oxygen demand after t = 160 min.

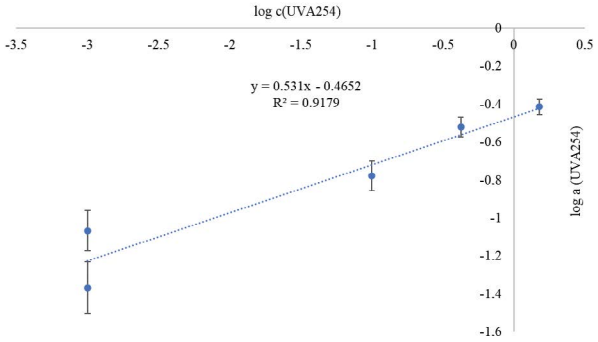

Fig. 21. Relationship between the logarithm of adsorbent concentration on activated carbon surface and in the solution, for leachate UVA(254) after t = 160 min.

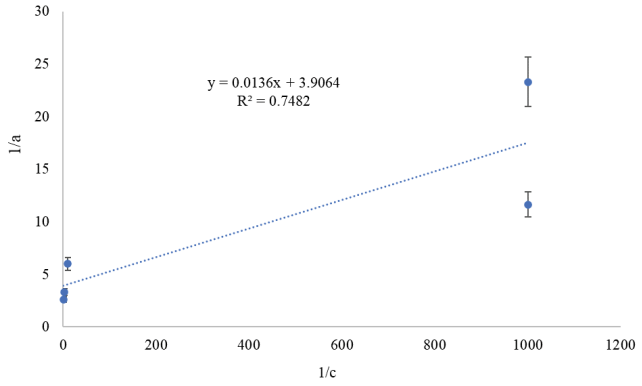

Fig. 22. Relationship between the converse of adsorbent concentration on activated carbon surface and in the solution, for leachate UVA(254) after t = 160 min.

reduction from 2,442 to 21 mg·L^{-1}, which corresponds to 99.14%. Similar results were obtained for a time of 40 min, namely, 23 mg·L^{-1} and significantly worse for 10 and 20 min, namely, 43 and 42 mg·L^{-1}, respectively, which provides a colour elimination effectiveness above 98.2%. When using shungite as an adsorbent, the highest efficiency was recorded for D = 80 g·L^{-1} and an exposure time equal to 80 min. This corresponds to an effectiveness of approximately 80.5%, which is significantly less than for activated carbon, but still a promising result, impossible to be achieved by the 4 other sorbents within this experiment. In the case of D = 80 g·L^{-1} of zeolite and the time t = 80 min, the obtained treated leachate colour equalled 751 mg·L^{-1}, which corresponds to an elimination of 69.25%. Shortening the remediation time and reducing the dose results in the leachate colour increasing to a value above 1,000 mg·L^{-1}. The effect appears already at a dose D = 40 g·L^{-1} and for the time of 80 min. The adsorption process may involve bonding of heavy metals by the solid phase. In the course of the conducted experiment, a significant change in the concentration of Cd, Cu, Cr, Ni, Pb, Zn and Co was recorded. The highest efficiency was obtained for an exposure to a sorbent dose D = 80 g·L^{-1} for Cd – 99.99%; Cu – 86.37%; Cr – 93.46%; Ni – 77.96%; Pb – 82.04%; Zn – 57.62% and Co – 98.39%. The results of these tests are shown in Table 5. Detho et al. [22] have discovered that zeolites, which have hydrophobic properties and are effective in the ion exchange, will be perfect at the removal of heavy metals. It was proven by the research conducted by Latosińska and Czapik [31], who used sewage sludge ash modified by the temperature and NaOH for the treatment of leachates (pH 8.03, COD 3,834 mg·L^{-1}, colour 5,929 mg·Pt·L^{-1}, TOC 1,688 mg·L^{-1}). They achieved a high degree of the removal of copper (90%, reaction time 10 min, dose 0.1 g·L^{-1}) and lead (100%, reaction time 10 min, dose 0.1 g·L^{-1}) as well as organic compounds. Satisfactory results of the reduction of lead were obtained also for sewage sludge ash, which had not undergone modification (96%, reaction time 20 min, dose 0.1 g·L^{-1}).

Figs. 15–22 show selected adsorption isotherms for the landfill leachate remediation process, obtained on the basis of the conducted experiment. Average or low R^2 determination coefficients were obtained in a majority of cases, which proves poor match between experimental data to Freundlich and Langmuir predictors. An exception is the leachate colour and UVA(254) elimination process with activated carbon (Figs. 16 and 22). This proves an extremely complex sorption mechanism in the landfill leachate-solid matrix system.

4. Conclusions

Based on the conducted tests, the results demonstrated an excellent effectiveness of activated carbon in removing leachate colour and UV adsorbance, as well as good effectiveness in eliminating the COD over a broad range of doses and a wide process duration spectrum. The highest efficiency in this respect was also obtained for shungite, and slightly lower effectiveness values for zeolite. It was impossible in this case to satisfactorily adsorb the substances responsible for a high value of the UVA(254) adsorbance index in landfill leachates. In consequence, UVA(254) elimination effectiveness for zeolite reaches 14.5% maximum. In this respect, zeolite effectiveness is significantly behind the two sorbents above, however, it is clearly superior to sewage sludge, spent coffee grounds and walnut shells. Laboratory tests did not confirm satisfactory effectiveness of these adsorbents. Walnut shells are particularly disappointing. When added to a solution as an adsorbent, they lead to a clear growth in UV absorbance. The greatest effect of the removal of the COD (13.5% for sewage sludge and t = 10 and 20 min; 19.9% for walnut shells and t = 160 min; 12.9% for spent coffee grounds and t = 10 min) and the colour (34.9% for sewage sludge and t = 10; 26.6% for walnut shells and t = 20 min) for alternative sorbents were obtained for dose 5 g·L^{-1}. Higher doses and longer time cause the deterioration of the initial parameters of landfill leachates. Far better effects of the removal of the COD were obtained for shungite (76% for D = 80 g·L^{-1} and t = 160 min), activated carbon (75.5% for D = 80 g·L^{-1} and t = 10 min) and zeolite (57.5% for D = 40 g·L^{-1} and t = 80 min). In the aspect of the elimination of UVA(254), the greatest results were obtained for activated carbon (99.97%, D = 40 g·L^{-1}, t = 160 min), shungite (74%, D = 80 g·L^{-1}, t = 40 min) and zeolite (14.5%, D = 80 g·L^{-1}, t = 160 min). In the aspect of the elimination of the colour, the greatest results were obtained for activated carbon (99.14%, D = 80 g·L^{-1}, t = 60 min). With the use of shungite as the adsorbent, the greatest result (80.5%) was obtained for D = 80 g·L^{-1} and t = 80 min. For zeolite (69.25%) at D = 80 g·L^{-1}, t = 80 min The research allowed

to discover a significant change of the concentrations of the analysed chemical elements – the greatest results were obtained at the exposition to the following doses of sorbents D = 80 g·L^{-1} for Cd – 99.99%; Cu – 86.37%; Cr – 93.46%; Ni – 77.96%; Pb – 82.04%; Zn – 57.62% and for Co – 98.39%. However, it seems that a properly performed conditioning of sewage sludge, coffee grounds or walnut shells may significantly improve the effectiveness of these matrices in terms of landfill leachate remediation processes in the future. This will enable the application of relatively inexpensive waste substances. The research in this field will be continued.

Acknowledgements

The research was carried out at the Solid Biomass and Biogas Energy Laboratory, which is part of the "Cenwis – Scientific and Implementation Center for Smart Specializations of the Świętokrzyskie Region" Contract No. RPSW.01.01.00-26-0001/17, 2017-2021

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