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Experimental investigation of the plant sorbents in comparison with Fibroil usable for runoff cleaning from petroleum products

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The capability of organic (plant) sorbents: Club-rush and Lesser Bulrush (from the lake shore) for removal of petroleum products (PP) from runoff was investigated. Dynamic process was investigated using filters packed with these materials. The third material analysed, synthetic sorbent Fibroil, was chosen for comparing the results. The laboratory experiments were performed in two stages, with the following being filtrated: tap water with a diesel admixture, road runoff contaminated with PP, and also suspended solids. The significance of this work is due to high runoff filtering rate (~10 m/h) and high PP concentrations in the runoff (>50 mg/L) used in the experiment. In these cases the use of sorbents is limited. *Schoenoplectus lacustris* and *Typha angustifolia* fillers are quite efficient (92–98%) in PP removal from runoff at a 10 m/h filtering rate. Use of them for filtration was possible longer than Fibroil, it clogs up quickly. *Schoenoplectus lacustris* and *Typha angustifolia* are a natural source for the production of PP sorbents. The results obtained in this experimental work can be used in the design of equipment for the treatment of PP contaminated runoff.

Key words: stormwater runoff, petroleum products, filtration, plant sorbents

INTRODUCTION

Road runoff produced from gas stations and car parking areas is one of the major sources that contribute oil and grease to receiving waters (Thomson et al., 1997; German, Svensson, 2005). Mostly gas stations have oil–water separators, oil may escape from the system due to peak flows during severe storm events (Khan et al., 2004). Once oil comes in contact with water, it forms emulsion that needs to be treated before it is disposed, because of toxic and hazardous properties of its components (Aberg et al., 2006; Brown, Peake, 2006; Ignatavičius et al., 2009; Saito, 2010). Petroleum products (PP) concentration in the runoff collected from highways and motorways usually reaches about 50 mg/L (in exceptional cases even higher PP concentrations, reaching 400 mg/L, may be observed) (Khan et al., 2004; Muhammad et al., 2004). Considering negative road runoff effect on aquatic ecosystems, it is clear that discharge of such runoff into surface water bodies is dangerous, as PP concentration often exceeds the allowable 5 mg/L limit by ten times. The negative effects on ecosystems and long-term effects of environmental pollution caused by PP contaminants require the development of a wide range of technologies for removing oil from soil and water (Deschamps et al., 2003; Muhammad et al., 2004; Cambiella et al., 2006; Nolde, 2007; Fuerhacker et al., 2011).

Road runoff produced from petrol stations and car parking areas is usually treated by precipitate tanks and filters (Muhammad et al., 2004; Mažeikienė et al., 2005; Maiti et al., 2011). In reality, the flow of storm water is uneven, constantly changing the concentration of run-off pollutants.

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These reasons cause enlargement of conventional technologies, extend time duration of technological processes and also complicate designing of new facilities (Lim, Huang, 2007; Ibrahim et al., 2009; Sobgaida et al., 2010). When sorbents are in use, special attention should be paid to the initial mechanical cleaning of stormwater and filtering rate (Rahmah, Abdullah, 2010; Rengasamy et al., 2011). To ensure efficient use of absorptive materials, it is necessary to evaluate stormwater contamination by suspended solids or turbidity by adjusting filtered runoff content properly, otherwise sorbent acts as a mechanical filter (Deschamps et al., 2003). One method of removing oil in gas station runoff is to equip the existing drainage systems or catch basins with an insert that captures PP. Inserts are the devices installed at the inlet of the stormwater catch basins to reduce pollutants (Mažeikienė et al., 2005; Sobgaida et al., 2010).

There are several types of inserts that rely on synthetic sorbents for contaminant removal. Commercial sorbents are most commonly used and also there are synthetic sorbents made of polypropylene or polyurethane (German, Svensson, 2005). The oil and grease removal performance of these sorbents is promising but they are expensive. Currently more and more publications appear about the using of natural organic sorbents for run-off treatment, as they are more acceptable than synthetic sorbents from the environmental point of view (Hiroshi et al., 2009; Abdullah et al., 2010; Asha, Thiruvenkatachari, 2010; Ali et al., 2011). Different organic materials have been investigated for oil sorption from water or run-off such as peat moss, vegetable fibres and their composites, sawdust, sisal fibre (*Agave sisalana*), coconut fibre (*Cocos nucifera*), vegetable-sponge (*Luffa cylindrica*), silk threads, mucor (*Mucor rouxii*), and kapok fibre (*Ceiba pendatra*). Sorption filter fillers made from these materials are 42–98% efficient in removing oil products (Moriwaki et al., 2009; Rahmah, Abdullah, 2010; Srinivasan, Viraraghavan, 2010; Wang et al., 2012; Svegl et al., 2013; Sanchez-Galvan et al., 2013). Although organic sorbents are less efficient in oil removal, they should not be underestimated. The costs associated with most biomass sorbents are only for their collection and preparation because the sorbents are available in local areas and can be obtained for free (Lim, Huang, 2007; Sanchez-Galvan et al., 2013; Svegl et al., 2013). Some of sorbents are aquatic weeds, which decrease aesthetics and interfere with waterway navigation. Utilizing these materials as PP sorbents will provide a side advantage on the removal of these last materials from water environment. The aim of this study was to investigate the abilities of organic (plant) sorbents – *Schoenoplectus lacustris* and *Typha angustifolia* – growing on the lakesides to remove petroleum products from runoff when it flows through filters filled with these materials.

MATERIALS AND METHODS

Sorbents

Three sorbing materials were chosen for the experiment: *Schoenoplectus lacustris*, *Typha angustifolia* and synthetic sorbent Fibroil for comparison.

The first investigated material was Club-rush (Lat. *Schoenoplectus lacustris*), a species of the *Cyperaceae* plant family. *S. lacustris* is a tall rhizomatous perennial herb of standing or flowing fresh water, in conditions ranging from eutrophic and base-rich to oligotrophic and base-poor. It occurs in ponds, lakes, canals, dykes and slow-moving rivers, usually in water 0.3–1.5 m deep. For the experiment described in this paper, Club-rush was taken from Kaimynai Lake in Lithuania. Long Club-rush stems (collected during the winter) were cut into 0.5–1.0 or 1.0–1.5 cm pieces before placing them into the filter column in the laboratory (Fig. 1).

The second investigated material was *Typha angustifolia*. *Typha* spp. (Lesser Bulrush) are aquatic macrophytes often regarded as weeds due to their ability to form dense monospecific stands which reduce the biodiversity of wetland systems and clog drainage ways. For the experiment described in this paper, Lesser Bulrush was taken from wet-

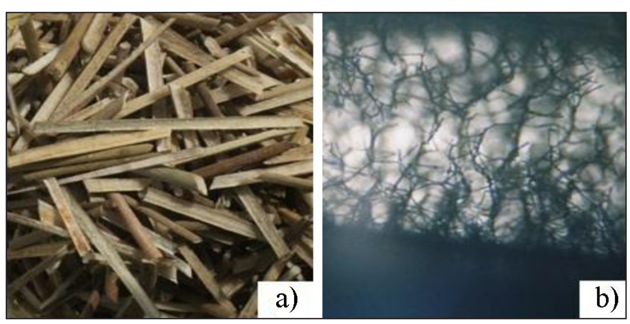

Fig. 1. Searching sorbing material *Schoenoplectus lacustris*: a) pieces before placing them into the filter column; b) structure of stem layer (at magnification of 100×)

lands of Lithuania. Lesser Bulrush stems (collected during the winter) were cut into 0.5–1.0 or 1.0–1.5 cm pieces before placing them into the filter column in the laboratory (Fig. 2).

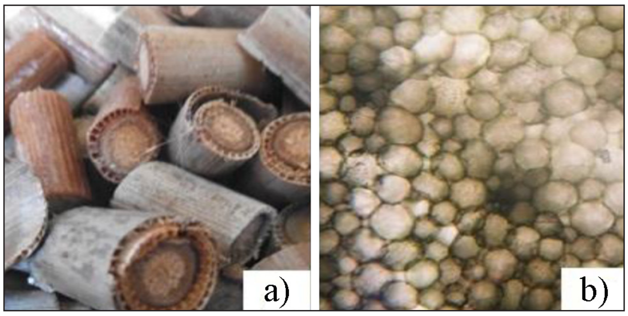

Fig. 2. Searching sorbing material *Typha angustifolia*: a) pieces before placing them into the filter column; b) structure of steam layer (at magnification of 100×)

The third material – a synthetic sorbent Fibroil – was chosen for comparison of the results. Sorbent material Fibroil was invented in the Czech Republic for capturing or separation of oil and other products from mixtures containing water. Fibroil test results show that this sorbent has many advantages: it can be used 10–15 times (sorbent capabilities decrease 50%), used sorbent requires no special treatment – it can be burnt in furnace, sorbent effectively absorbs oil products (properties as indicated by the producer). FIBROIL® is water-proof, thus it can be used in filter fillings for filtering of oil products and mixtures containing water. A photo of Fibroil is given in Fig. 3.

Fig. 3. Synthetic sorbent Fibroil

Sorption in batch tank

Sorption in batch tank was carried out with 1 g sorbent material. Sorbents were placed in 500 mL Erlenmeyer flask with 150 mL of oily wastewater, with PP concentration 4 550 mg/L and pH 6.8. Sample was then mixed in laboratory stirrer for 30, 60, 90 min at room temperature (22 ± 1 °C). Then sorbents were taken out of flask and drained. Sorbent efficiency was determined by the use of an extractive-gravimetric method. PP sorption capacity (g/g) for experiments in batch tank was determined according to the formula:

$$q = C_i - C_f , \quad (1)$$

where: C_i = the initial PP concentration (g/L); C_f = the final PP concentration (g/L).

Measurements were repeated 3–4 times. So at present only average concentration is presented in this paper. Average concentration was determined by the formula:

$$\bar{c} = \frac{1}{n}\sum_{i=1}^{k} c_i m_i , \quad (2)$$

where: c_i = concentration of substances at typical points, m_i = probability at recurrence of concentration; n = number of measurements; k – number of different values of concentration.

Constructed filter system

A pilot test equipment was constructed in the chemical laboratory of Water Management Department. Filter model scheme is given in Fig. 4.

Road runoff or tap water was poured into the tank (1). The pump (12) carried it to the tank (2), where a stable liquid capacity was maintained, ensuring a steady flow velocity in the pipe (4) at an incline of 3°. The velocity was controlled by opening the valve in line with the filtrate flow. The velocity was measured every 10 minutes. Diesel from the vessel was inserted by peristaltic pump (3) into the running liquid at a rate at which the initial oil concentration (P_i) was reached before going through the sieve at the end of the pipe, where samples were taken. Afterwards, road runoff or tap water with initial PP concentration (mixtures) was provided at an equal load through the sieve to the filter column (6) with a cross-sectional area of 0.006 m^2. The mixtures were filtrated through a 15 or 20 cm high filler layer (weight and density of layer presented in Table 2) as these height values are usually used in oil separators. Filtering column was filled with searching sorbent material. Filtering rate of 10 m/h was chosen for the experiment. Mixture samples

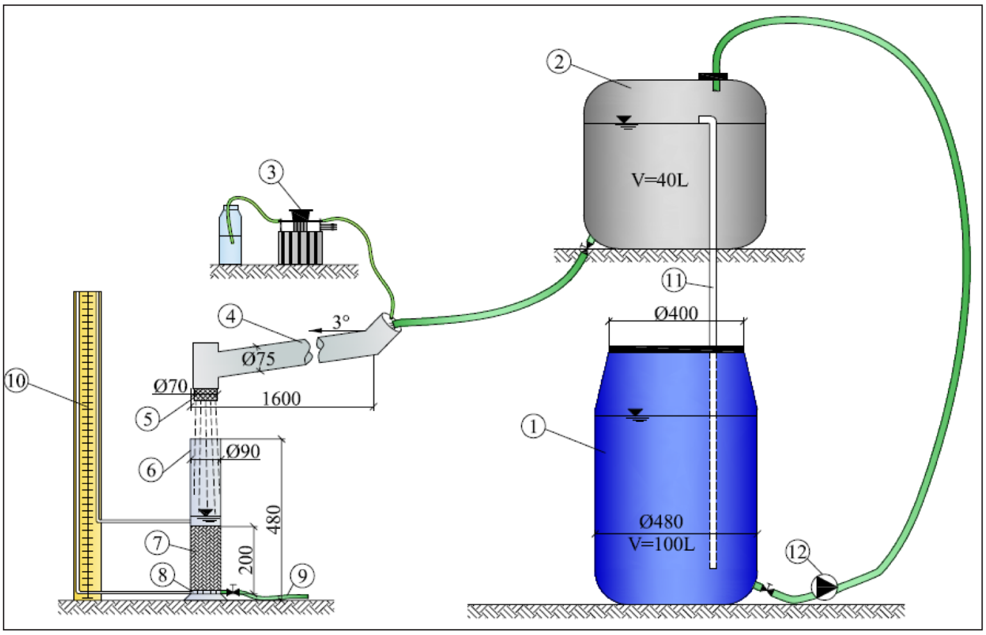

Fig. 4. Filtration setup: (1) 100 litres runoff tank; (2) 50 litres tank; (3) pump for diesel dosing; (4) pipe that provides run-off with inserted PP to the filter; (5) sieve; (6) filter column; (7) filter filler; (8) rubble layer supporting the filler; (9) flexible hose for filtrate samples; (10) piezometer; (11) overflow pipe; (12) pump

were put in jars with a 0.5 L capacity before putting it in the filter column every ten minutes. Filtrate samples from the hose (9) were put in jars with a 0.5 L capacity too. Contamination in these samples was measured by estimating PP and suspended solids concentrations. Pressure losses in the filter column were measured by a piezometer (10).

Two types of mixtures were used for experimental research.

First type mixture was prepared by mixing water from water supply network (concentration of sediments (TSS) was around 0.5–1 mg/L) with diesel fuel (2 class (CS51), standard: LST EN 590-2009), second type mixture was prepared by mixing natural surface runoff water with diesel fuel.

Every single filtration experiment was made with new sorbing filling material. The results were calculated using formulas shown below.

PP amount (mg) in runoff before the filter was calculated according to the formula:

$$P_{inf} = \sum_{i=1}^{n} P_i \cdot Q_i \cdot \Delta t_i, \quad (3)$$

where: P_i = PP concentration before the filter at each time interval (mg/L); Q_i = water flow rate at each time interval (L/min); Δt_i = interval time of sampling (min).

Suspension in filling layer PP (mg) is calculated with the formula:

$$P_{susp.} = \sum_{i=1}^{n} (P_i - F_i) \cdot Q_i \cdot \Delta t_i, \quad (4)$$

where: F_i = PP concentration in the filtrate at each time interval (mg/L).

PP removal from runoff efficiency (%):

$$\eta = \frac{P_{sorb}}{P_{inf}} \cdot 100, \quad (5)$$

Measurements of the filtering parameters were repeated 3–4 times, average concentration was determined by the formula (2).

Errors consisted of specimens taking and measuring tools accuracy errors. After the experimental work, a statistical analysis of the results was prepared by eliminating unreliable values above 95% confidence interval.

At the beginning of experiments, inside of chemical laboratory of Water Management Department (VGTU), runoff water was measured for concentration of sediments. Average concentration was determined by the formula (2). Sediments concentration was measured by filtering specimens through glass fibre filters, and drying sediments at temperature 105 ± 2 °C (LST EN 872:2005) until stable weight. Temperature and pH were measured by WTW gauge pH 325-B/SET-2. Photographs of steam structure were made by microscope "MOTIC" B1223A (magnification up to 1 000× photo ability). Specimens PP concentration was measured at Ecological supervision laboratory, UAB "Grinda". PP (nonpolar and slightly polar carbohydrate, extracted in hexane) concentration was determined by ISO 9377-2:2002 standards. More than 1 000 mg/L concentration of PP was determined by weight method (LAND 90-2010).

RESULTS AND DISCUSSION

Results of investigations of runoff samples are presented in Table 1:

Table 1. **The pollution of road runoff samples taken from the Geležinio Vilko Street Tunnel**

Sample taking points	[PP] (mg/L)	[TSS] (mg/L)	pH
1.	32.9	345	7.3
2.	25.6	230	7.4
3.	106.5	420	7.0
4.	19.7	95	7.4
5.*	4550	1580	6.8

* Sample was taken from glacial thaw swamp

Sorption in batch tank was carried out with road runoff samples from 5 points (Table 1) in which a considerable PP concentration occurred. Sorption capacities of *S. lacustris, T. angustifolia* and Fibroil for real oily runoff in batch tank were 1.5 g/g, 1.3 g/g and 7 g/g. Fibroil showed higher sorption capacity compared to plant sorbents. Obtained sorption capacity of Fibroil is lower in comparison with literature data: 14 g/g (Absorption..., 2008). Lower sorption efficiency for real oily runoff samples coud be due to the form of PP present in water. Real runoff occurred in the form of stable emulsion.

From the results obtained in batch system, the idea was to develop the efficiency of investigated sorbents for emulsified PP in experimental filtration equipment. The experiments began with an aquatic mixture (tap water with diesel admixture) filtration test in order to avoid the effects of suspended solids usually existing in the runoff. A filtering rate of 10 m/h was chosen and maintained. Table 2 shows the overall results of the experiments with aquatic mixture.

A comparison of results for different sorbents used for PP removal show that higher PP removal from aquatic mixtures efficiency was obtained by using Fibroil as filter filler. However, the obtained PP removal efficiency for plant sorbents is good in comparison with literature data (Hiroshi et al., 2009; Abdullah et al., 2010; Asha, Thiruvenkatachari, 2010; Ali et al., 2011; Srinivasan, Viraraghavan, 2010). PP concentration increase before the filter (P_{inf}) showed an increase in the amount of suspended PP in the filling layer. The efficiency of *S. lacustris* layer to contain PP was higher than that of Fibroil because P_{inf} in test 1 was larger than in test 10.

However, it is importantly PP concentration in the filtrate (F_i); it was 122 mg/L (initial) and 579 mg/L (last) (Table 2, test 1). In reality, such a

Table 2. **Summarized data of the experiments with an aquatic mixture**

Test	Filtration term, min	Size of pieces, cm	Height of layer, cm	Weight of layer, g	Density of layer, g/cm³	P_{inf}, mg/L		F_i, mg/L		η, %
						initial	last	initial	last	
Schoenoplectus lacustris										
1.	120	1.0–1.5	10	40.0	0.06	1512	5359	122	579	64
2.	80	0.5–1.0	15	80.0	0.08	77	119	8	17	87
3.	250	0.5–1.0	15	84.2	0.09	25	212	2.6	14	91
4.	80	0.5–1.0	15	44.3	0.05	46	98	2.8	3	98
5.	360	0.5–1.0	20	70.6	0.06	2082	4580	61	102	97
Typha angustifolia										
6.	80	1.0–1.5	15	87.0	0.09	98	135	9	17	87
7.	100	1.0–1.5	15	77.0	0.08	154	600	13	25	93
8.	80	0.5–1.0*	15	61.48	0.06	25	212	2.6	14	91
Fibroil										
9.	80	–	10	30.5	0.03	200	368	0.2	3	99.2
10.	360	–	20	61.0	0.05	158	426	0.5	5	99.0

large PP concentration cannot be in the natural runoff. Lithuanian legislative documents limit PP concentration in runoff. The highest allowable average concentration of PP in flow must be ≤5 mg/L (instantaneous concentration – ≤7 mg/L). Therefore further experiments were carried out with real road runoff contaminated by PP and also suspended solids and they continued till $F_i \leq 7$ mg/L. Assays of road runoff contained the same initial contamination (TSS and PP). In order to improve oil removal efficiency, the filler height was increased from 10 cm to 20 cm. At the beginning of experiments, some pressure losses inside filtering cylinder appeared, which depended on sorbent material structure and density of filtering column filings. After some time, the increase of pressure loss depended on the characteristics of mixtures being filtered (PP products and sediments concentrations). It was possible to keep a stable filtering rate until pressure losses reached 25 cm, after that rate (10 m/h) started to decrease. When the pressure losses increased, the filter fillers became impermeable and the experiment was ended. The results of the filtration experiments are shown in Fig. 5 and Fig. 6.

During the experiments, when the filtration time was longer, the initial PP concentration in the water increased before it reached the filter. As the filter gradually clogged, the filtering rate decreased, but diesel was provided at the same flow rate as it had been set.

This is the reason for a higher PP concentration in the measuring point before reaching the filter. From Fig. 5 one can see that the filtration through the plant fillers lasted 360 min, while filtration through Fibroil filler lasted only 160 min. Fibroil filler clogged first, unless the PP removal from water efficiency using Fibroil filler was high (99%).

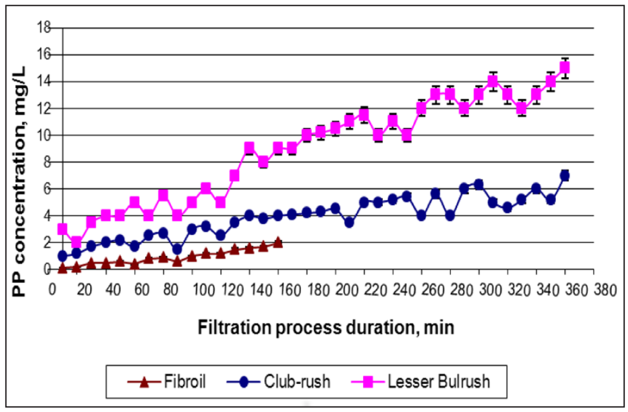

Fig. 6. PP concentration in filtrate (flow rate: 10 m/h, t: 22 °C, P_{inf}: 100–150 mg/L, TSS: 32 mg/L)

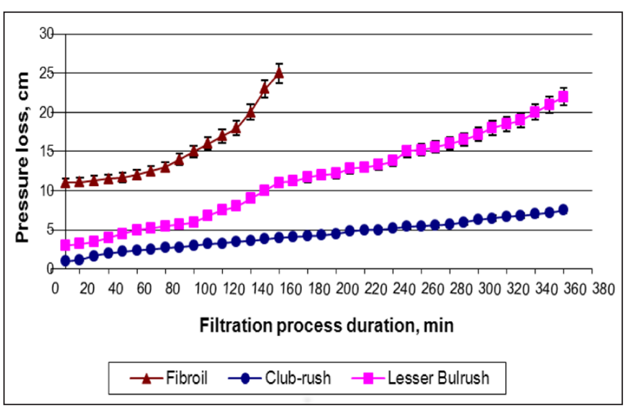

Fig. 5. Pressure loss inside fillings (flow rate: 10 m/h, t: 22 °C, P_{inf}: 100–150 mg/L, TSS: 32 mg/L)

Using Lesser Bulrush filler in the filter, a PP concentration of 7 mg/L as regulated by the law was reached after two hours of filtration. However, this concentration was reached only after six filtration hours with using Club-rush. The effectiveness of PP removal (on average) from mixtures of the two plant fillers was good (97–98% for *S. lacustris* and 91–92% for *T. angustifolia*). In comparison with the first (with aquatic mixtures) experiment, the Club-rush filler removed PP from the assays of road runoff more efficiently due to its height 20 cm (as it was two times higher). Lesser Bulrush also removed PP more efficiently (efficiency increased from 87 to 92%). After the evaluation of all the results, it is presumed that Club-rush and Lesser Bulrush fillers can be used in different structures for PP removal from runoff or sullage. Their efficiency of PP removal from water or runoff in dynamic mode is similar or even better than it is in case of other natural organic sorbents: rise husk: 73%; wood chip: 74%; kapok fiber: 83% (Khan et al., 2004).

CONCLUSIONS

The paper analyses the ability of Club-rush and Lesser Bulrush growing on lakesides for removal of PP from runoff when it flows through filters filled with these materials. The results indicated that plant sorbents – *Schoenoplectus lacustris* and *Typha angustifolia* – could be used for treatment of water and removal of PP. The significance of this work is due to high runoff filtering rate and high PP concentrations in the runoff (>50 mg/L) used in the experiment. In these cases the use of sorbents is limited. Club-rush and Lesser Bulrush fillers are quite efficient (92–98%) in PP removal from runoff at a filtering rate of 10 m/h. It was possible to keep stable filtering rate until pressure losses reached 25 cm, after that rate (10 m/h) started to decrease. Filtration through the plant fillers lasted 360 min, while filtration through Fibroil filler lasted only 160 min. Fibroil filler clogged first, unless the PP removal from runoff was high (99%). Using Lesser Bulrush filler in the filter a PP concentration of 7 mg/L as regulated (instantaneous) by the law was reached after two hours of filtration. However, this concentration was reached only after six filtration hours with using Club-rush. The results obtained in this experimental work can be used in designing equipment for the treatment of PP-contaminated runoff from ship terminals, oil bases, petrol stations as well as sullage from roads and tunnels.

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NAFTOS PRODUKTUS IŠ PAVIRŠINIŲ NUOTEKŲ ŠALINANČIO FILTRO SU AUGALINIAIS SORBENTAIS EKSPERIMENTINIAI TYRIMAI

Santrauka

Šiame straipsnyje aprašomų eksperimentų tikslas – ištirti organinių (augalinių) sorbentų – ežerinio meldo ir siauralapio švendro – tinkamumą šalinti naftos produktus (NP) iš paviršinių nuotekų, kai nuotekos filtruojamos pro filtrus, užpildytus šiomis medžiagomis. Siekiant palyginti rezultatus, eksperimentams pasirinkta trečioji medžiaga – sintetinis sorbentas Fibroil. Laboratoriniai eksperimentai vykdyti keliais etapais: buvo filtruojamas vandens ir dyzelino mišinys bei nuo kelio paviršiaus surinktos nuotekos, užterštos NP ir skendinčiosiomis medžiagomis. Šis darbas reikšmingas tuo, kad nuotekos filtruotos dideliu greičiu (~10 m/h) ir jose buvo didelės (>50 mg/l) NP koncentracijos. Tokiais atvejais sorbentų naudojimas yra ribotas. Augalų *Schoenoplectus lacustris* ir *Typha angustifolia* užpildai gana efektyviai (92–98 %) šalino NP iš nuotekų, kai filtravimas vyko 10 m/h greičiu. Juos galima ilgiau naudoti nuotekų filtravimui nei Fibroilį, kuris greitai kimšosi. *Schoenoplectus lacustris* ir *Typha angustifolia* yra gamtinis NP sorbentų šaltinis. Šio eksperimentinio darbo rezultatai gali būti pritaikyti projektuojant įrenginius, skirtus naftos produktams išvalyti iš paviršinių nuotekų.

Raktažodžiai: paviršinės nuotekos, naftos produktai, filtravimas, augaliniai sorbentai