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OIL REMOVAL FROM WATER BY FILTRATION

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Abstract. Oil-contaminated water is most commonly treated with sorbent materials. In this experimental study, a fibrous sorbent material Fibroil was used. The experiment was carried out with tap water and clarified stormwater. It was determined that the retention of contaminants is worse at high flow rates, which reduces the efficiency of treatment. Sorbent materials retain suspended solids and reduce water turbidity; thus, the water must be clarified and pretreated before it is supplied to the sorption fillings. For the efficient use of sorbent material properties, the concentration of suspended solids in water supplied to the filter must be below 20 mg/L, while water turbidity must be below 15 NTU and the flow rate must be below 20 m/h. If the pressure loss in the sorption filler increases to 25 cm, it can be predicted that the oil concentration after treatment would exceed permissible environmental requirements (5 mg/L). The derived sorption and hydraulic properties of the material can be used to evaluate the efficiency of existing operative stormwater treatment plants as well as to design new facilities.

Keywords: water, oil, sorbent, water pollution, suspended solids, filtration, Fibroil.

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Introduction

Due to increasing industrial activities, more and more contaminants from urban areas such as suspended solids, heavy metals, oil products, etc., reach the environment via stormwater (Pitt *et al.* 2002; German, Svensson 2005; Mimi 2008). Oil is one of materials that pollute the earth entrails (Pitt *et al.* 2002; Deschamps *et al.* 2003; Ke *et al.* 2005). Commonly used in many human activities, oil inevitably comes into contact with water; and this pollution presents a threat to the surface waters and drinking water sources (Browne *et al.* 2008; Paulauskienė *et al.* 2009). Oil spilt in the environment undergoes a wide variety of weathering processes, which include evaporation, dissolution, dispersion, photochemical oxidation, microbial degradation, adsorption onto suspended materials, agglomeration, etc. (Wei *et al.* 2003; Khan *et al.* 2004; Husein *et al.* 2008). Oil concentration in stormwater collected from highways and motorways often reaches 50 mg/L. In some cases, such as the first summer rain, oil concentration can even reach 400 mg/L (Muhammad *et al.* 2004; Khan *et al.* 2004). Stormwater must be treated before its release into the surface waters and pretreated before it is discharged into stormwater networks. The reduction of oil spread in the environment and of the further pollution is one of the most important ecological objectives (Nolde 2007; Zhu *et al.* 2010; Shrestha, Brodie 2011).

According to the Lithuanian legislation, the allowed concentrations of oil and suspended solids (SS) in discharges into stormwater networks are the following: the annual average SS concentration of 150 mg/L (the highest instantaneous concentration of 300 mg/L) and the annual average oil concentration of 10 mg/L (the highest instantaneous concentration of 30 mg/L). Whereas, when the stormwater is released directly to the natural environment, the allowed annual average SS concentration is 30 mg/L (the highest instantaneous concentration of 50 mg/L) and the annual average oil concentration of 5 mg/L (the highest instantaneous concentration of 7 mg/L).

Many methods and facilities are used for stormwater treatment, for example, oil separators, ponds, plant

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treatment, infiltration, filters or hydrodynamic separators (Herrmann, Schmida 2000; Davies-Colley, Smith 2001; Garg *et al.* 2004; German, Svensson 2005). Meanwhile, to separate oil from water, different methods such as sedimentation, filtration, sorption and flotation are used (Herrmann, Schmida 2000; Shin, Chase 2005; Shrestha, Brodie 2011).

The European standards LST EN 858-1:2002 and LST EN 858-2:2003 regulate the principles of design, testing and marking of oil separators. There are two classes of separators according to treatment efficiency: the class I (oil concentration in effluent does not exceed 5 mg/L) and the class II (concentration up to 100 mg/L). However, for example, in Belarus and Russia, the required treatment level is 0.5 mg of oil per litre. The requirements for oil effluent into environment may be stricter than 5 mg/L, depending on local conditions. In order to achieve a higher efficiency of stormwater treatment, it is necessary to install sorption fillers. The filters shorten oil separation time; therefore, the size of equipment can be reduced.

The application of sorption processes in removal of pollutants from stormwater is widely analysed in literature (Deschamps *et al.* 2003; Garg *et al.* 2004; Mažeikienė *et al.* 2005; Branvall *et al.* 2006). Used sorbents can be classified as polymers, natural materials or treated cellulosic materials. Most commonly used commercial sorbents are synthetic sorbents made of polypropylene or polyurethane. They have good hydrophobic and oleophilic properties (Deschamps *et al.* 2003). The literature provides a lot of information on the application of different sorbents for oil removal from water surface, i.e. collection of oil from aqueous solutions in a static way (Deschamps *et al.* 2003; Wei *et al.* 2003; Annunciado *et al.* 2005; Husein *et al.* 2008). Different studies of sorbents were carried out at low flow rate of 0.3–0.4 m/h, (Rajakovic *et al.* 2006) or 0.5–1.5 m/h (Rahmah, Abdulah 2010). However, there are no studies of sorbents in dynamic mode, at high flow rates and high oil concentrations.

Using sorbents, particular attention should be paid to primary mechanical stormwater pretreatment and flow rate. In order to achieve an efficient use of absorption materials, it is necessary to estimate the quantity of suspended solids in stormwater or its turbidity, so that the sorbent doesn't operate as a mechanical filter (Davies-Colley, Smith 2001; Rahmah, Abdullah 2010; Pitt *et al.* 2002; Garg *et al.* 2004).

1. Materials and methods

Hydrophobic sorbent material under the brand name Fibroil is often used for oil separation filters. This material can be fibrous or of cloth type. Chemical composition of the material: approx. 90% of polypropylene and 10% of limestone, density of 50–80 kg/m^3, the ignition temperature is 450 °C, under static conditions 1 gram of the cloth absorbs approx. 18 grams of oil. The parameters of filtration through Fibroil fillings are not specified in any literature. Nevertheless, Fibroil is the most commonly used sorbent in the Baltic countries. The sorbent can be used from 10 to 15 times, although its sorption properties decrease by approx. 50%. Its regeneration is achieved by mechanical wring out. The aim of this study was to evaluate the treatment efficiency using fibrous Fibroil material, depending on flow rate and concentration of oil and suspended solids.

In the dynamic filtration study, it is essential to ensure a constant flow rate and uniform initial contamination. In the experimental work, a method of continuous oil insertion into the water was used. The pumps supplied water and oil to the filtration equipment at a constant flow rate. This method enables to prepare mixtures of any concentration. The study was carried out with the initial oil concentrations of 50 mg/L, 100 mg/L and 150 mg/L. The scheme of the experimental setup is shown in the Fig. 1.

Two types of water were used for the experiment. The base solution was prepared by supplying the tap water with diesel fuel (2 class (CS51), standard LST EN 590-2009). The other type was prepared from stormwater brought from the industrial district of Vilnius, where separate stormwater network was constructed. To achieve a more exact simulation of stormwater treatment in real facilities, the stormwater was precipitated for 2 hours before filtering.

Water was poured into the tank (1). Then, the pump carried it to the tank (2), where a stable water level was

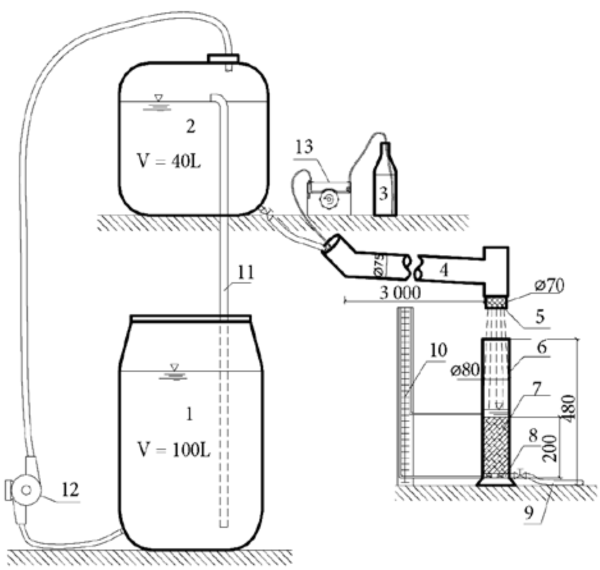

Fig. 1. Experimental setup: 1 – 100 litre tank; 2 – 50 litre tank; 3 – diesel tank; 4 – pipe for water and diesel mixing; 5 – sieve; 6 – filter column; 7 – Fibroil; 8 – support layer; 9 – flexible hose for samples; 10 – piezometer; 11 – overflow pipe; 12 – pump; 13 – pump for diesel dosing

maintained, which guaranteed a constant flow rate by pipe (4) with the incline of 0.003 as well as mixing of water and diesel. The quantity of running water was controlled by a valve. The flow rate of filtrate was measured every 10 minutes. A peristaltic pump inserted diesel from the diesel tank (3) to the running water at a rate permitting to achieve designed oil concentration behind the sieve and equal distribution of water and diesel.

Then, water and oil were supplied at a steady flow rate to the filter column (6) with a cross sectional area of 0.005 m^2. It was filtrated through the filter layer of 20 cm in height. The total amount of the sorbent material used for one test amounted to 76 grams (70 kg/m^3 filling weight). The flow rate chosen was 20 m/h and 30 m/h. Every 10 minutes samples of water and treated water were taken, and the concentration of oil and suspended solids as well as turbidity were measured. Pressure losses were measured by a piezometer (10).

Every single experiment was conducted with a new filling of the same weight and density. Fibrous sorbent material was used for filtration. The filtration experiments were repeated three times for reliability of results. Estimated potential errors of samples and analysis methods are presented in the graphs. Water was analysed using standard methods: suspended solids (SS) (LST EN 872:2000), turbidity (LST EN ISO 7027:2002) and total petroleum hydrocarbons (TPH) (ISO 9377-2:2000).

2. Results and discussion

The experiments with tap water and oil were repeated several times as the quantity of water was not limited (more than 1.2 m^3 for one experiment). However, real stormwater transportation to the laboratory always has a limited scope. The tests were limited due to transportation of stormwater to the laboratory.

First, a hydraulic test was conducted. Pressure losses were measured in the sorption filter while clean tap water was supplied at rates of 20 m/h and 30 m/h. A linear dependence of pressure losses on a flow rate was determined. After 10 minutes from the beginning of the filtration process, the pressure losses reached 5.2 cm at 20 m/h rate and 10.5 cm at 30 m/h rate, respectively. Later, pressure losses gradually increased by 0.5 cm every hour. The quality of the water used in the experiment is presented in Table 1. Some of the tests were carried out with tap water and others – with real clarified stormwater.

The following abbreviations are used in the paper: W stands for tests with tap water, and W1–W5 for tests with clarified stormwater.

The experiments showed that once water was mixed with diesel, water turbidity and suspended solids concentration increased. For example, when 150 mg of oil was added to 1 litre of tap water (W), the turbidity increased to 32 NTU, and suspended solids concentration – to 7 mg/L.

Table 1. Pollution of examined water

Water types (abbreviations)	Initial water pollution		
	SS (mg/L)	Turbidity (NTU)	Oil (mg/L)
Tap water (W)	0.5	1.0	0
Water (W1)	20	30	2.0
Water (W2)	8	10	1.5
Water (W3)	25	34	2.3
Water (W4)	40	60	5.5
Water (W5)	10	15	2.0

The amount of oil supplied to the filter was calculated by the formula:

$$Oil_{in} = \sum_{i=1}^{n} C_i \cdot Q_i \cdot \Delta t_i, mg, \quad (1)$$

where: C_i – oil concentration before the filter at each time interval, mg/L; Q_i – water flow rate at each time interval, L/min; Δt_i – sample taking time interval, min.

The amount of retention oil was calculated:

$$Oil_{retention} = \sum_{i=1}^{n} \left(C_i - C_t\right) \cdot Q_i \cdot \Delta t_i, mg, \quad (2)$$

where: C_t – oil concentration in the filtrate at each time interval, mg/L.

The efficiency of oil removal from water:

$$\eta = \frac{Oil_{retention}}{Oil_{in}} \cdot 100, \%. \quad (3)$$

After the experimental work, a statistical analysis of obtained results was prepared by eliminating unreliable values above the limit of 95% of the confidence interval. Summarised experiment data is shown in the Table 2.

Fig. 2 provides the results of water filtration through Fibroil filling. The filtration of water with initial oil concentration of 50 mg/L took 6 hours (1.2 m^3 of mixture were filtrated) until pressure losses increased up to 25 cm. It was not possible to continue mixture filtration at the

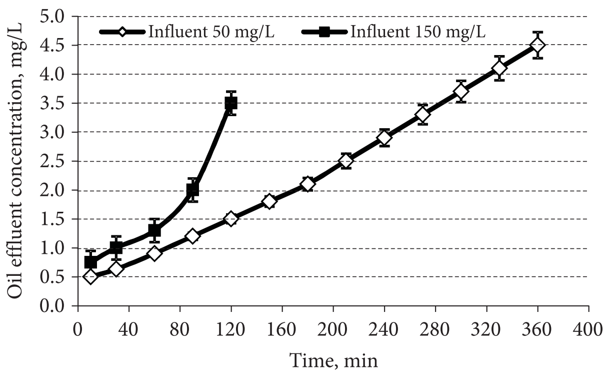

Fig. 2. Results of water treatment at the flow rate of 30 m/h

Table 2. Results of oil removal from different water types

Water types	Initial concentration, mg/L			Effluent concentration, mg/L			Supplied amount of oil, g	Oil retention	
	Min.	Ave.	Max.	Min.	Ave.	Max.		Total amount, g	η, %
Tap water (W)	47.5	50	52.5	0.5	2.6	4.7	57.6	54.8	95.1
	142.5	150	157.5	0.7	2.8	4.8	57.8	56.7	98.1
Water (W1)	47.9	52.5	57.1	0.3	2.3	4.3	14.4	13.8	95.8
Water (W2)	48.3	50.5	52.7	0.8	2.5	4.2	34.7	33.0	95.1
	145.4	151.4	157.4	0.6	3.6	6.5	53.3	52.0	97.6
	47.5*	51.5*	55.5*	0.2*	1.5*	2.8*	46.1*	45.7*	99.1*
Water (W3)	49.8	52.6	55.4	0.3	2.2	4.1	15.1	14.5	96.0
	151.4	153.5	155.5	2.5	3.7	4.9	29.5	28.8	97.6
Water (W4)**	95.5	127	158.5	0.4	1.7	2.9	21.8	21.5	98.6
Water (W5) **	51.7	69.3	86.9	0.3	2.3	4.3	13.2	12.7	97.3

*Filtrated at constant rate of 20 m/h, **Filtrated at inconstant rate.

rate of 30 m/h due to clogging of the filter. Oil concentration in the filtrate increased from 0.5 mg/L to 4.7 mg/L. During 6 hours of filtration, 54.8 g of oil were accumulated using 76 g of sorbent filling. The filtration of water with initial oil concentration of 150 mg/L lasted for 120 minutes until pressure losses reached 25 cm. At the end of filtration, there were 3.7 mg/L of oil in the sample. The higher was the water pollution, the shorter was the filter operating time.

Fig. 3 shows the increase of pressure losses during the filtration of tap water and clarified stormwater at the initial oil concentration of 50 mg/L and flow rate of 30 m/h. It was ascertained that the experiments with tap water did not reflect the situation in real oil separators as filtration of real clarified stormwater was 4 times shorter. Oil removal experiments with tap water were not reliable; nevertheless, the majority of experiments described in references were conducted with tap water. After this evaluation, further experiments were conducted only with clarified stormwater.

The results showed that constant rate could only be maintained if pressure losses in sorbent filling did not exceed 25 cm. As pressure losses increased, the flow rate decreased and the filtration processes were interrupted. The increase of pressure losses is mostly influenced by water contamination by suspended solids, as the oil concentration in both tested water types was the same. The value of pressure losses in the filter could be increased by elevating filter column. However, it was not necessary because in practice, when pressure losses increase up to 20–25 cm, emergency water level in tanks is reached, and the stormwater is usually directed to the bypass.

An experiment was carried out with clarified stormwater (W2). Water pollution before the experiment is presented in Table 1. The water was treated at the initial oil concentrations of 50 mg/L and 150 mg/L and at the flow rates of 20 m/h and 30 m/h. The main results are provided in Figs 4 and 5.

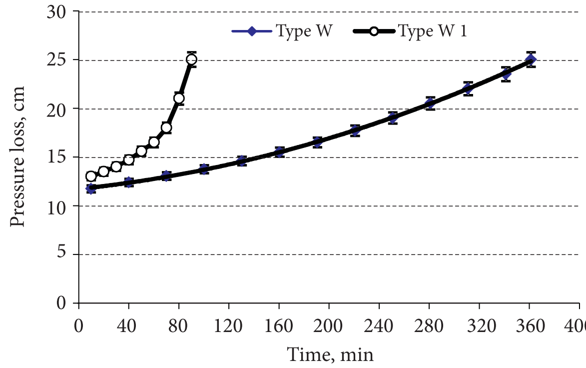

Fig. 3. Pressure losses in the filtration of tap water (W) and clarified stormwater (W1)

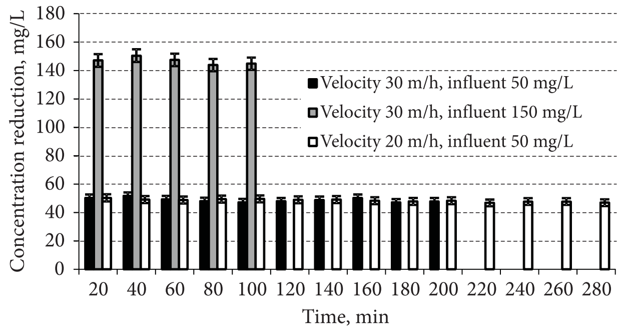

Fig. 4. Quantity of oil removed from clarified stormwater (W2)

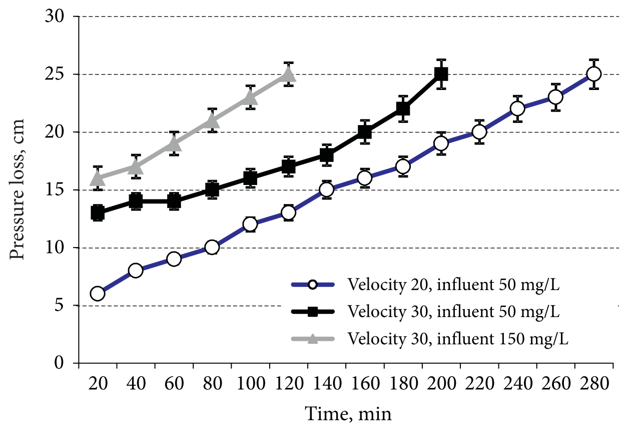

Fig. 5. Graphics showing pressure losses according to oil concentration and flow rate (clarified stormwater W2)

Once water was filtrated at the rate of 30 m/h (until pressure losses reached 25 cm), the efficiency of oil retention varied from 98.3 to 92.7% (with the oil concentration of 50 mg/L) and from 99.5 to 97.6% (with the oil concentration of 150 mg/L). The retention efficiency was higher at a higher initial oil concentration; still, due to greater amount of diesel, the pressure losses grew faster. After 100 minutes from the beginning of filtration, the oil concentration in the filtrate exceeded the limit of 5 mg/L. The retention efficiency in water with 50 mg/L of oil was higher at the filtration rate of 20 m/h than of 30 m/h. The pressure losses in the filter reached the critical limit 1.4 times faster at the filtration rate of 30 m/h than at 20 m/h. Therefore, the oil retention efficiency of sorbent is influenced by flow rate (the lower is the rate, the higher is the efficiency) and initial oil concentration (the higher concentration, the higher efficiency). The duration of the filtration process was influenced by stormwater contamination depending on its turbidity and quantity of suspended solids (Fig. 4, Fig. 6). When these indicators were higher, the pressure losses reached the critical limit faster. It was observed that the turbidity of filtrate samples didn't exceed 4 NTU during the experiment, and the concentration of suspended solids didn't exceed 2–3 mg/L. These measurements confirm that the sorbent cannot only retain oil products but other pollutants as well.

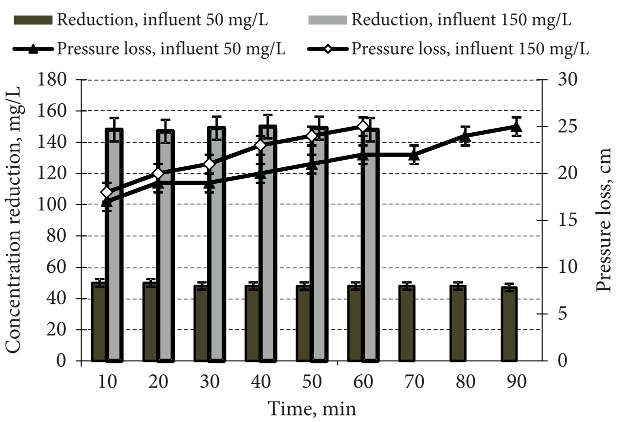

Fig. 6. Quantity of oil removed from clarified stormwater (W3)

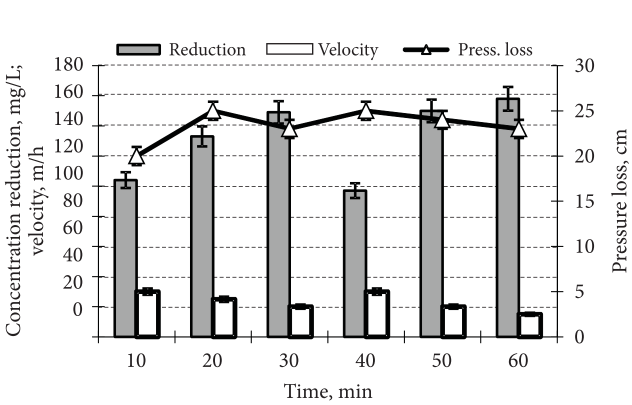

Fig. 7. Quantity of oil removed from clarified stormwater (W4)

The highest retention efficiency was achieved in the filtration of the most contaminated clarified stormwater (W4). The parameters of water contamination after the insertion of diesel were as follow: the turbidity 69 NTU, the SS concentration 46 mg/L and the oil concentration approx. 130 mg/L. It was not possible to maintain a constant flow rate at such a high water contamination degree because of clogging of the filter filling and the sieve (Fig. 7). In 30 minutes, the flow rate declined from 30 m/h to 20 m/h. Diesel was supplied at a constant rate. As the flow rate decreased, the oil concentration before filter increased and reached 149 mg/L at the 30th minute of filtration. While the pressure losses were growing, the flow rate as well as the oil concentration decreased (from 1.5 to 0.4 mg/L). After 30 minutes of filtration, the sieve got blocked and the filtration process was terminated. When the sieve was cleaned, the filtration continued at the flow rate of 30 m/h. The oil concentration in the filtrate was around 0.5 mg/L. However, after 10 minutes of filtration, the flow rate decreased to 20 m/h again, and, respectively, the oil concentration in filtrate samples went down to 0.4 mg/L. Afterwards, the flow rate continued to decrease due to fast clogging of the filter, and the filtration process was stopped.

Clogging of the sorption filling during filtration of clarified stormwater was higher than during filtration of tap water, with the differences ranging from 3 to 5 times. Sorption filler not only retained oil, but also – suspended solids, solid and colloidal particles and other materials that water may contain. Clarity of treated water was approx. 2 times higher than clarity of the water under treatment. In order to increase the efficiency of the filter, stormwater must be clarified and pretreated on coalescence modules. Otherwise, Fibroil filling would partly operate as a mechanic filter and lose its technological properties in a short period of time. The use of Fibroil as a mechanical filter is not beneficial technologically or economically. It is recommended to supply the filter with water that has the concentration of suspended solids below 20 mg/L and the turbidity up to 15 NTU. To ensure these conditions, the pretreatment of stormwater in settlement tanks and coalescence filters must be efficient. In order to evaluate the operating experience of existing stormwater treatment plants, it is necessary to eliminate undissolved oil particles getting into the filter filling. The oil separators must be designed in such a way that with the growth of pressure losses and water level in the tank exceeding 25 cm, the alarm must switch on and the stormwater is automatically directed to the bypass. Further increase of water level would cause flooding of the inflow pipe and the flow distribution unit. The study proved that the highest flow rate through the fibrous Fibroil filter should not exceed 20 m/h, as more intense flow produces higher pressure losses and reduces efficiency of treatment and sorption.

The sorbent material properties declared by the producer and the experimental research results show that the maximum permissible hydraulic losses in the filter filling appear before the sorbent material is saturated with oil.

Fig. 8 provides dependence of the pressure losses on suspended solids concentration and filtering time. When suspended solids concentration in the water under treatment increased from 8 mg/L to 40 mg/L, the filtering time in the sorption filter decreased by 10 times. An analogous situation must be found in real treatment plants. Thus, the control of pressure losses or, to be precise, of water level in the tank, could be one of the most important factors in the optimisation of operational parameters of oil separators.

Before using any sorbent materials in dynamic mode (when water is filtered through the material), it is necessary to not only estimate its sorption capacity but also – mechanical filtration properties. However, these data are not generally provided in the data sheets of the sorbent materials. It was found that the filter retention efficiency depends on the flow rate and turbidity as well as on the concentration of suspended solids. In all tested cases, the pressure losses reached 25 cm before the effluent oil concentration came up to 5 mg/L (as required by Lithuanian environmental standards). This shows that the pressure loss in the filter fillings is one of the most important and limiting factors for the design of filters with sorbent fillings and it might be used as easily determinable technological criteria.

Fig. 9 presents the dependence of oil retention on suspended solids concentration. The graph shows that when the quantity of suspended solids in the water under treatment is higher, the retention filter capacity is lower.

It is probable that the filter would be able to retain the same amount of oil if the stormwater is filtered more times at a lower rate. Still, under real conditions, the flow rate cannot decrease too much compared to the design value, as the treatment plants would be less efficient than planned.

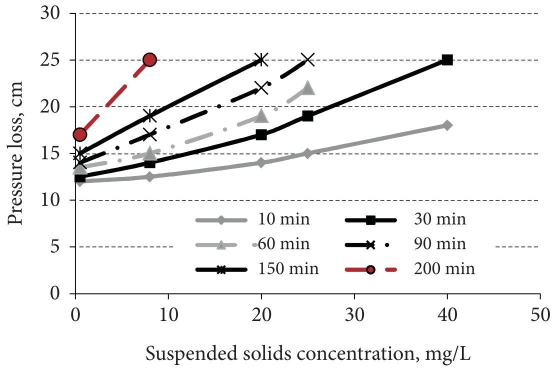

Fig. 8. Dependence of pressure losses on suspended solids concentration and filtering time

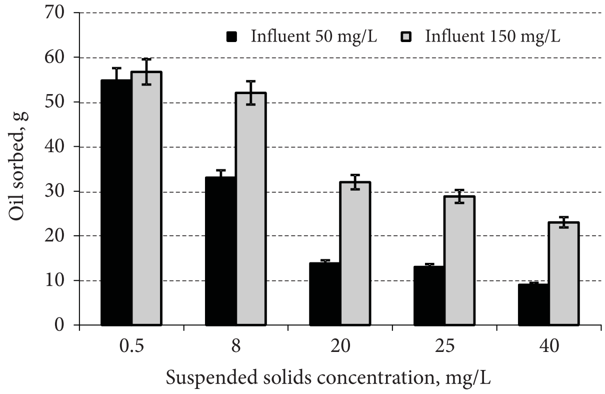

Fig. 9. Dependence of oil retention on suspended solids concentration

Conclusions

1. The treatment efficiency using Fibroil fibrous material depends on the flow rate, turbidity and suspended solids concentration. Taking into account the treatment efficiency, the recommended flow rate must be below 20 m/h.

2. Sorbent materials retain suspended solids and reduce water turbidity; therefore, the water must be clarified and pretreated before supplying to the sorption fillings. It is recommended to feed the Fibroil filling with water that has suspended solids concentration below 20 mg/L and turbidity below 15 NTU.

3. When the pressure losses in the sorption filter filling increase up to 25 cm, it can be predicted that the oil concentration after treatment will exceed permissible environmental requirements (5 mg/L). When stormwater treatment plants are designed, it must be evaluated that the pressure losses should not exceed 25 cm.

4. Before the use of sorbent materials in oil separators and other devices, it is necessary to estimate its retention and hydraulic properties.

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