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EFFICIENCY OF PETROLEUM PRODUCTS REMOVAL FROM AQUEOUS MIXTURES WITH DIFFERENT SORBENT MATERIALS

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Abstract. The aim of this paper was to investigate the performance parameters and efficiency of different sorbent materials (FIBROIL® and DUCK) for oil removal from aqueous mixtures and natural surface runoff water in dynamic mode. An experimental study was carried out. In all cases, pressure losses (in both sorbents filling) reached 25 cm point before petroleum products concentration of filtrate specimens reached 5 mg/L. The influence on filtering effectiveness had the filtering rate (the slower, the better effectiveness) and initial petroleum products concentration (the bigger inside research boundries, the better effectiveness). Both sorbent materials removed petroleum products from mixtures effecively (95,0 % – 99,7 %). DUCK was a little bit more effective than FIBROIL®, but it becomes foul more quickly.

Keywords: Sorbent material, FIBROIL®, DUCK, oil, petroleum products removal, filtration.

1. Introduction

With expansion of the oil business the level of environmental pollution risk is inevitably increasing. Oily water contamination from industries has been a major problem due to the effects on housing wastewater runoff. When oil comes in contact with water, it forms oil-in-water emulsion or floating film that needs to be removed before it is discharged into the environment (Deschamps *et al.* 2003; Cambiella et al. 2006; Ju *et al.* 2008). Even very low concentrations of oils can be toxic to microorganisms responsible for biodegradation in conventional sewage processes. Lithuanian anti-pollution standards limit concentration of PP in run-off which can to get into natural environment. The most allowable average concentration of PP in flow must to be ≤ 5 mg/l (instantaneous concentration – ≤ 7 mg/l).

Several physical approaches (plate sedimentation, thickener, floatation chamber, centrifugation and deep-bed filtration, have been developed to tackle the problem (Muhammad *at al.* 2004). The use of various sorbents is the most common technique to treat floating oil on water surface because of the possibility of its collection and reuse (Annunciado *et al*. 2005; Baltrėnas and Branvall 2006; Branvall *et al.* 2006; Huang and Lim 2006; Lim and Huang 2007). The sorbents are categorized into three distinct groups, namely organic natural sorbent, organic synthetic sorbent and inorganic mineral sorbent. Most available oil sorbents in the market are organic synthetic-based sorbent (Rajakovic *et al.* 2006; Lim and Huang 2007). Nowadays, for economic reasons, organic and inorganic natural products, such as rice straw, corn cob, peat moss, wool, cotton, cotton grass, cattail fiber, rice husk, sawdust, wood chips barks, bagasse, milkweed, zeolite, clinoptilolite, vermiculite are now becoming increasingly important as an alternative to the synthetic sorbents (Mažeikienė *et al.* 2008; Husein *et al.* 2008; Abdullah *et al.* 2010; Rahmah and Abdullah 2010).

In this paper, two types of sorbents for oil removal from wastewater were investigated: organic synthetic sorbent FIBROIL^R and inorganic mineral sorbent DUCK. The aim of this study was to analyze and compare their performance and efficiency. It is well established that some synthesic sorbents have high oil sorption capacity. On the other hand, minerals are considered to be very efficient and cheap sorbents due to their chemical and mechanical stability, high surface area and structural properties. DUCK is a powdery hydrophobic expanded mineral, showing a unique morphology. High porosity and peculiar characteristics make him very efficient sorbent for removal of PP from water. However, DUCK was tested only in static conditions. In this paper were investigated its properties of sorption in dynamic mode.

The oil sorbents can also be used as coalescing media in a fibrous or granular bed to treat stable oil-in-water emulsion (Rajakovic *et al.* 2006). However, the treatment of oily wastewater by sorption and coalescence is expensive because of the high costs of the oil sorbents. Such a use can be an alternative for these wastes instead of disposal as it can provide cheap sorbent for oily wastewater treatment.

2. Object and techniques

A pilot test equipment was constructed at the Water Management Department‘s chemical laboratory. Filter model‘s scheme is given in Figure 1.

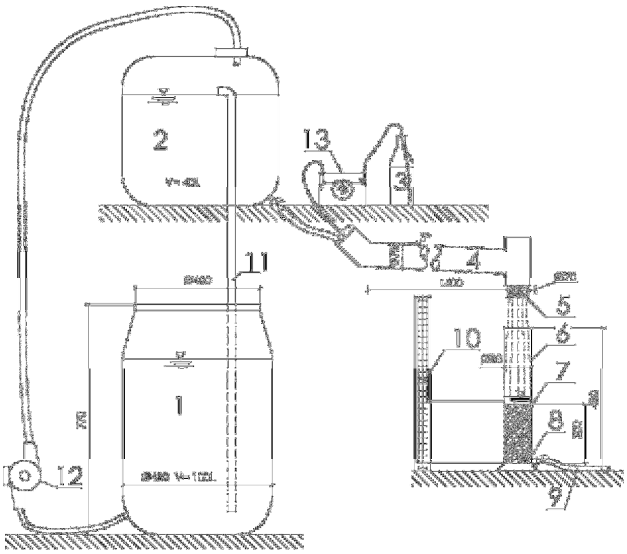

Fig 1. Pilot test equipment: 1 – 100 l tank for water (runoff water); 2 – 50 l plastic container; 3 – bottle filled with diesel fuel; 4 – supply pipe with flow regulation grid; 5 – percolator; 6 – filtering column; 7 – FIBROIL^R or DUCK filing; 8 – rubble layer to keep filling in place; 9 – sampling pipe; 10 – piezometer; 11 – pipe for excessive water amount; 12 – pump; 13 – pump for dosing diesel

Two types of mixtures were used for experimental research. I-st type mixture was prepared by mixing water-supply water (turbidity around 1 NDV) with diesel fuel (2 class(CS51), standard: LST EN 590-2009), with oil products concentration in water before filter – 50; 100; 150 mg/L. II-nd type mixture was prepared by mixing natural surface runoff water with diesel fuel.

Runoff water or water-supply water was poured into 100 L capacity tank (1); pump (12) took it to the plastic container (2). Then water got to the supply pipe (with incline of 3°) with flow regulation grid. Flow rate was measured every 10 minutes. Pump (13) from bottle filled with diesel fuel (3) poured PP (diesel fuel) into flowing fluid at a beginning of a pipe at exact rate, that at the end of a pipe, behind percolator to run up the initial C_0 concentration of oil (petroleum) products (specimens were taken here). Afterwards runoff or water-supply water with initial PP concentration, with equal load achieved by percolator, were introduced to filtering column (6) cross-section area of 0,005 m^2, and was filtrated through 20 cm height, 76 grams (60 kg/m^3 filling weight) FIBROIL®, or same height (108 grams, 0,6-2,0 mm size granules) DUCK layer. Filtering rate was 15-30 m/h.

Filtering column was filled with searching sorbent material. Sorbent material FIBROIL® was invented in Czech republic, for capturing or separation of oil and other products from mixtures containing water. FIBROIL® test results shown that this sorbent has many advantages: it can be used 10-15 times (sorbent capabilities decreases 50 %), fast and easy regeneration – by mechanical wring out; used sorbent requires no special treatment – it can be burned in furnace; sorbent effectively absorbs oil products (Absorption material FIBROIL®). FIBROIL® is water-proof, that’s why it can be used in filter fillings for filtering of oil products and mixtures containing water. FIBROIL®’s photo is given in Figure 2.

Fig 2. Sorption material FIBROIL®

Scientific literature does not indicate parameters for filtering with FIBROIL®. Together FIBROIL® is fairly expensive material, so it was important to investigate how runoff water characteristics influence PP sorbtion effectiveness.

Sorption material DUCK is used to collect liquid, lighter than water products from water surface. DUCK’s photo is given in Figure 3.

Fig 3. Sorption material DUCK

It is made from natural vulcanic mineral. It is light, selective, hydrofobic. 1 kg of sorption material DUCK can to absorb around 3,98 L of diesel fuel (Sorbentas…2010). Usually powdered over areas of leakage, and collected afterwards (after absorbing PP). Its capabilities of removing PP from runoff water in dynamic mode has not been researched.

Filtrate specimens and specimens of mixtures I and II (before filtration cylinder) were taken every ten minutes, and placed into 0,5 L jars. Pressure losses that occurred inside of filtering cylinder were measured by piezometer (10). Every single filtration experiment was done with new FIBROIL® or DUCK media, which had exact same weight and density. Measurements of the filtering parametes were repeated 3-4 times. Errors consisted specimen taking and measuring tools accuracy errors.

In the beginning of experiments, inside of VGTU Water Management Department's chemical laboratory runoff water was measured for concentration of sediments and turbidity. Sediment concentration was measured by filtering specimens through membrane filters, and drying sediments in 105 °C temperature until stable weight. To measure turbidity *Spectroquant* NOVA 60, from MERCK company was used (turbidity was determined in nefelometric feculence units).

Specimens PP concentration was measured at UAB "Grinda" Ecological supervision laboratory. This laboratory is certificated by Republic of Lithuania Environmental Protection Agency. Total oil products (non polar and slightly polar carbohydrate, extracted in hexane) concentration was determined by ISO 9377-2:2000 standards. Device used: oil analyzer AN-2 (measuring accuracy (0,04 ÷ 1000) mg/l ±2%).

3. Experimental results

In the beginning of experiments, some pressure losses inside filtering cylinder were appeared, which depended on sorbent material structure, density of filtering column filings and rate of filtering. Initial pressure loss dependence on filtering rate (while filtering I type mixture with initial 50 mg\L PP concentration) is shown in figure 4.

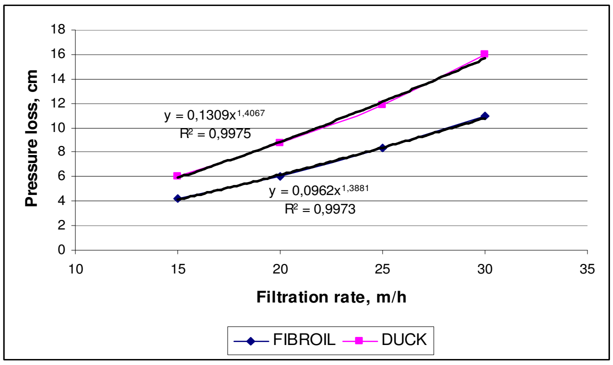

Fig 4. Dependence of initial pressure losses inside fillings on filtering rate

After some time, pressure loss increase depended on characteristics of mixtures being filtered (oil products and sediments concentrations). It was possible to keep stable filtering rate until pressure losses reached 25 cm, after that rate started to decrease.

As it is shown in figure, over first ten minutes of filtering, the increase of pressure losses was directly dependent on filtering rate: when filtering rate was 15 m/h, pressure losses were 4,2 cm; when filtering rate was 30 m/h, pressure losses were 11,0 cm (in FIBROIL® filling) and 6,0 cm (30 m/h) – 16,0 cm (15 m/h) (in DUCK filling).

The increase of pressure losses inside FIBROIL® and DUCK fillings while filtering I and II type mixtures (with initial PP concentration C_0 = 50 mg/L) at the rate of 30 m/h are shown in figure 5.

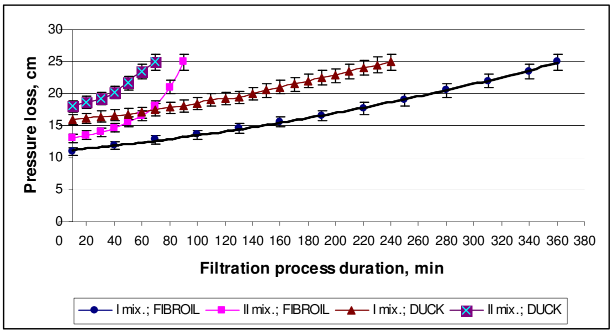

Fig 5. Dependence of pressure loss increase on type of water mixture

Turbidity of I type mixture inside tank 1 (Fig1) was 1 FDV (sediments concentration – 0,5mg/L), and II type mixture was 30 FDV (sediments concentration – 20mg\L). After pouring diesel fuel before FIBROIL® filling, both mixtures had PP concentration of 50 mg/L. Pouring diesel fuel caused increase of turbidity and sediment on both of mixtures: I mixture turbidity increased to 6 FDV (sediments concentration – 2 mg/L), II mixtures turbidity increased to 37 FDV (sediments concentration – 23 mg/L). As it is shown in figure, that pressure losses increase during filtering was smaller in case of mixture type I than type II: pressure losses increased to 25 cm point after 360 minutes (in FIBROIL® filling), 240 minutes (in DUCK filling) of filtering, when in case of type II mixture it reached the same point after 90 minutes (in FIBROIL® filling), 70 minutes (in DUCK filling) of filtering. So, pressure losses increase was influenced by impurity of mixtures (of turbidity and sediments), even know PP concentration was equal in both mixtures.

Table 1 represents the effectiveness of PP removal (on average) from mixtures.

The results of filtering mixture type I at rate of 30 m/h are shown in figure 6. Water turbidity inside tank was 1 FDV, sediments concentration – 0,5 mg/L. After pouring diesel fuel to achieve PP concentration of 50 mg/L, mixtures turbidity increased to 6 FDV, and sediments to 2 mg/L. After increasing PP concentration to 150 mg/L, mixtures turbidity reached 32 FDV, sediments concentration – 6,5 mg/L.

Table 1. PP removal from mixtures effectiveness

Measurement time, min	v, m/h	C_o, mg/L	C_f, mg/L	C_o-C_f, mg/L	Effectiveness, %
FIBROIL					
	30	50,0			
Initially	30	50,0	0,5	49,5	99,0
Finally, 360	30	52,2	4,5	47,7	91,4
	30	150,0			
Initially	30	150,5	0,75	149,8	99,5
Finally, 120	30	150,0	3,5	146,5	97,7
	15	50,0			
Initially	15	51,0	0,3	50,7	99,4
Finally, 700	15	50,5	2,5	48,0	95,0
	15	150,0			
Initially	15	150,4	0,5	149,9	99,7
Finally, 220	15	151,6	3,4	148,2	97,8
DUCK					
	30	50,0			
Initially	30	51,5	0,3	51,2	99,4
Finally, 240	30	52,0	2,7	49,3	94,8
	30	150,0			
Initially	30	151,0	0,55	150,45	99,6
Finally, 60	30	149,5	2,9	146,6	98,1
	15	50,0			
Initially	15	51,5	0,2	51,3	99,6
Finally, 470	15	51,0	1,8	49,2	96,5
	15	150,0			
Initially	15	150,5	0,4	150,1	99,7
Finally, 120	15	151,5	2,0	149,5	98,7

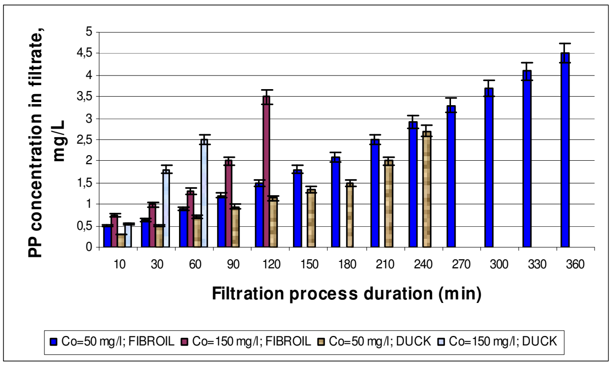

Fig 6. Results of filtering I type mixture (v = 30 m/h)

As it is shown in figure, filtration of I mixture through FIBROIL® filling proceeded 6 hours (1152 L of mixture being filtered), before pressure losses reached 25 cm point. Oil products concentration in filtrate specimens (C_f) changed from 0,5 mg/L to 4,5 mg/L, so it did not reached reglamented 5 mg/L concentration. Over 6 hours of filtering, 76 g FIBROIL® filling collected (absorbed) 54800 mg of oil products. Filtration of I mixture with initial PP concentration of 150 mg/L proceeded 120 minutes before pressure losses reached 25 cm point. At the end of filtering C_f was 3,4 mg/L, and also did not reach aforementioned concentration. Filtration of I mixture through DUCK filling proceeded 4 hours (when C_o was 50 mg/L) and 1 hour (when C_o was 150 mg/L), before pressure losses reached 25 cm point. Oil products concentration in filtrate specimens (C_f) changed from 0,3 mg/L to 2,9 mg/L, so it did not reached reglamented 5 mg/L concentration.

As can be seen from data shown in figures, every time after pouring diesel fuel the turbidity and sediments concentration increased. And dependent on these, duration of runoff water filtration decreased. In all cases, pressure losses reached 25 cm point before PP concentration of filtrate specimens reached 5 mg/L.

While using both sorbents, biggest sorbtion effectiveness was achieved filtering the most impure runoff water at the slowest rate. When filtration rate decreased from 30 m/h to 15 m/h, adsorbtion effectiveness increased, also influenced by time that runoff water spent inside sorbent filling (23 s, when v = 30 m/h; 33 s, when v = 20 m/h; 46 s, when v = 15 m/h). Subject to reduced filtration rate, duration of runoff water filtration increased.

A comparison of results for different sorbents used for PP removal show, that the better adsorbed quantity of PP was obtained by using DUCK as a filter medium. The highest PP removal efficiency (initially 99,7 %, finally 98,7 %) was achieved using DUCK filling: when the initial PP concentration in I type mixture was 150 mg/L (filtration rate was 15 m/h), the residual PP was (initially 0,4 mg/L, finally 2,0 mg/L) in filtrate. However by reason of quick blockage DUCK media proved unequal, than FIBROIL®.

4. Conslusions

1. During the laboratory experiment with a pilot test equipment, the possibility to use two different sorbent materials – FIBROIL® and DUCK – for the PP removal from water mixtures was shown.
2. Pressure loss increase dependencies from filtering rate were determined during the research. In all cases, pressure losses reached 25 cm point before PP concentration of filtrate specimens reached 5 mg/L.
3. After comparing results for both sorbent materials, it was revealed that DUCK initial pressure losses are bigger and previously reaches a point, that limits further filtration from maintaining a stable filtering rate (25 cm), than it is in FIBROIL® filling.
4. Filtering duration was influenced by impurity of mixtures from PP concentration and sediments. Mixtures that were less contaminated before filter (C_0 = 50 mg/L) were filtered for 240 minutes (through FIBROIL®); 60 minutes (through DUCK) more that, mixtures with bigger contamination (C_0 = 150 mg/L) at filtering rate of 30 m/h.
5. While filtering water mixtures (with initial PP concenration of 50 - 150 mg/L) influence on filtering effectiveness had both, filtering rate (the slower, the better effectiveness) and initial PP concentration (the bigger inside research boundries, the better effectiveness).

6. Both sorbent materials removed PP from mixtures effecively. DUCK was a little bit more effective than FIBROIL®, but it becomes foul more quickly.

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