Environmental Engineering and Management Journal

June 2019, Vol. 18, No. 6, 1193-1200 http://www.eemj.icpm.tuiasi.ro/; http://www.eemj.eu



"Gheorghe Asachi" Technical University of Iasi, Romania



VISCOSE-MAGHEMITE/GOETHITE POLYMERIC COMPOSITE AS SORBENT FOR OIL SPILL CLEANUP

Răzvan Rotaru, Maria Emiliana Fortună*, Corneliu Cojocaru, Petrișor Samoilă, Lucia Pricop, Valeria Harabagiu

"Petru Poni" Institute of Macromolecular Chemistry, 41A Gr. Ghica Voda Alley, Iasi, 700487, Romania

Abstract

The paper reports on the preparation of polysiloxane hydrophobized viscose-maghemite/goethite magnetic composite with properties appropriate for uses in oil spill sorption from polluted aqueous media. A multi-step procedure was employed to obtain the polymeric composite. The structures of the prepared samples and precursors were analyzed by Fourier transform infrared spectroscopy (FTIR), whereas their morphology was investigated by means of scanning electron microscopy (SEM). The hydrophobicity of the composite was confirmed by the water-contact-angle value of 93 degrees. The magnetic properties of the composite were evaluated showing a saturation magnetization (M_s) equal to 26.7 emu/g. The developed composite material unveiled relevant sorption capacities of 15-17 g/g for liquid hydrocarbons (decane/dodecane) and 35-37 g/g for motor oils uptake. The obtained results showed that the employed synthesis method led to a composite material with good magnetic and sorption properties, which can be applied as a potential oil spill sorbent in aquatic media.

Key words: ferromagnetic, oil spill, polymeric composite

Received: October, 2016; Revised final: March, 2017; Accepted: June, 2017; Published in final edited form: June 2019

1. Introduction

Oil is the most important fuel of our society and the main source for other products like synthetic plastics, lubricating oils, bitumen, saturated hydrocarbons, etc. Oil also represent a danger for aquatic medium due to possible leak in oceans during extraction and transportation (millions tons per year). Therefore, the clean-up of oil spillage is essential and is of current interest. A wide range of inorganic, organic or/and synthetic materials designed to remove spilled oil and other saturated hydrocarbons from contaminated waters are known (Abdul et al., 2012).

The ability of porous sorbents to remove oil spill is based on absorption and adsorption processes (Rotaru et al., 2014). Absorbents acts through capillary phenomena by collecting oils like sponges.

They are swelling materials enabling liquids to diffuse into their matrix. Adsorption is a surface phenomenon; it is rapid at the beginning and slows down near the equilibrium. Pollutants are only retained on the solid surface of the sorbent material. Usually, when porous or fibrous-porous sorbents are exploited for oil uptake both capillary and adsorption phenomena take place simultaneously (Rotaru et al., 2014). A good oil spill sorbent must present a great sorption capacity, good reuse potential and high hydrophobicity.

As regards cellulosic-based sorbents, these involves low-cost natural materials like kapok, sawdust, peat moss and others, which represent abundant and renewable resources (Cojocaru et al., 2011). Modified cellulose fibers were found many years ago to be able to absorb crude oil and saturated hydrocarbons (Johnson and Manjrekar, 1973).

^{*} Author to whom all correspondence should be addressed: e-mail: fortuna.maria@icmpp.ro; Phone: +40735511119

A literature survey indicated that the sorption capacity depends on the nature of sorbent and the type of oil pollutant. For instance, according to Husseien et al. (2008) the sorption capacity of the barley straw sorbent for gas oil uptake was equal to 8.5 g (oil)/g (sorbent). Suni et al. (2004) reported a value of 18 g/g for cotton grass (raw fibres) in mineral oil. Radetic et al. (2008) worked on recycled wool-based nonwoven material and determined a sorption capacity of 9.6-10.6 g/g for diesel oil uptake. Other authors (Senanurakwarkul et al., 2013) reported a sorption capacity of 10-12 g/g for recycled rayon waste-kapok mixtures in motor oil.

The adsorption of oils onto solid surface of materials occurs mainly due to physical attractive forces (i.e. Van der Waals forces with Debye, Kiesom and London dispersion energy components (Weber et al., 1991)). Thus, the adsorption of hydrophobic oils and hydrocarbons is closely related to the presence of hydrophobic groups onto the sorbent surface (Abdullah et al., 2010; Khan et al., 2004; Ribeiro et al., 2003; Srinivasan and Viraraghavan, 2010). The oil sorption occurs as a result of hydrophobic interaction between the sorbent and oily liquids (Annunciado et al., 2005; Likon et al., 2013; Ribeiro et al., 2003). Therefore the producing of hydrophobic sorbents is of practical interest.

Polysiloxanes are used as coating agents in the textile industry for hydrophobization of fibers and fabrics. Such treatment is based on siloxanes properties to diffuse on the surface of the textile support and to form hydrophobic films on the contact surface. Typically, polysiloxane copolymers containing Si-H functional groups are employed for this purpose, as they are able to easy undergo the cross-linking reaction leading to a hydrophobic and durable coating film, particularly in the presence of a suitable catalyst (e.g. Zn-octoate). Besides the very good effect of hydrophobization, such a film offers to the solid surface other properties, such as smooth and silky handle, water washing and solvent cleaning resistance, which are desirable properties for oil spill sorbent (Vazquez, 1999).

Another aspect is related to the practical use of the sorbents as a prompt action. In this regard, the development of the sorbents with magnetic properties is of real interest, especially to facilitate the separation between the spent sorbent and the aqua environment.

The goal of this work is to prepare and investigate new composite materials with magnetic properties as candidate sorbents for oil spill cleanup of aquatic media. In this respect, viscose-maghemitegoethite composites were prepared by means of the ultrasonic technique. To keep the fibrous structure of cellulosic material, the amount of magnetic material was selected to be less than 20 %. In addition, the composite was hydrophobized with a polysiloxane copolymer to enhance the water-repellent properties and to improve its applicability for oil spill cleanup. The oil sorption properties of the as prepared hydrophobic composite were compared with those of the pristine viscose fibers and of other reported oil sorbents. The easy magnetic recovery of the spent sorbent was also demonstrated.

2. Material and methods

2.1. Materials

Viscose fibers (Vs), (Lenzing AG, Austria; linear density, 1.3 dtex; fiber length, 39 mm), iron (II) sulfate heptahydrate (FeSO₄·7H₂O), decane (C₁₀H₂₂), dodecane (C₁₂H₂₆), purchased from Sigma-Aldrich, sodium hydroxide and toluene from Merck, zinc octoate as 16 % solution in mineral spirit (CSTex, Romanian product), Motor Oil Magnatec 5W-40 C3: Castrol, Motor Oil Superlub Universal 15W40: Mol-Lub Kft-UE (commercial products) were used as received. poly(hydromethyl-dimethyl)siloxane Α copolymer (PSi) of Mn = 1040 g/mol average number molecular weight and a weight polydispersity index 1.6 (synthesized according to a previously reported procedure (Pricop et al., 2005); active hydrogen content: 0.219 % (determined by modified Zerewitinoff method); IR (KBr pellets), cm⁻¹: 2853-2964 and 1413 (C-H); 2158 (Si-H); 1261, 845 and 800 (Si-CH₃); 1091-1032 (Si-O-Si)) was used a as hydrophobization reagent.

The appliances and devices engaged in the processes of preparation are as follows: ultrasound generator Sonics Vibracell (750W nominal electric power, 20 kHz ultrasound frequencies), vacuum oven Trade Raypa, Specac laboratory press, centrifuge EBA-21 Hettich.

2.2. Preparation of maghemite-goethite nanoparticles (MG)

MG nanoparticles were obtained according to a method previously described (Rotaru et al., 2017). In a typical procedure, $FeSO_4$ (1/2 w/w) and NaOH (1/4 w/w) aqueous solutions were prepared in ultrapure water and mixed (1/1 w/w ratio) in a Berzelius beaker, with a Sonics Vibracell ultrasound generator at 50 % from the maximum intensity. The beaker was placed on an ice bath and the temperature was rapidly increasing during sonication up to 25 °C. The energy dissipated in the ultrasonic bath was 109 kJ after sonication for 60 minutes. The sample was than centrifuged, washed with Milli-Q water, centrifuged again and dried in a Trade Raypa vacuum oven for 24 h at 40 °C, to obtain maghemite-goethite particles.

IR (KBr pellets), cm⁻¹: 885 and 795 (deformational modes of hydroxyl groups, δ -OH out of plane and in plane, typical for goethite); 579 cm⁻¹ (v Fe-O for γ -Fe₂O₃ phase) (Rotaru et al., 2017).

2.3. Preparation of viscose-maghemite-goethite composite (Vs-MG)

Vs-MG composite was obtained by ultrasonication of a 4/1 w/w Vs/MG suspension in distilled water for 15 min, in the Sonics Vibracell generator, at 50% from the maximum intensity.

During sonication, 29.3 kJ energy was dissipated into the mixture, and a temperature of 86 °C was reached at the end of the ultrasonication time span. The resulted Vs-MG fibers of a uniform brown color were dried in a vacuum oven at 40 °C, for 24 h.

2.4. Pressed viscose (Vsp)

The pressed viscose sample (Vsp) was prepared by using a Specac laboratory press. A pressing force of 10 tons on a surface of 1.327 cm^2 was applied to compact the Vs fibers.

2.5. Preparation of hydrophobic composite (Vs-MG-PSi)

Vs-MG-PSi hydrophobic composite was prepared by suspending 5 g Vs-MG composite in a 15 mL solution of 5% PSi concentration in toluene containing Zn octoate catalyst (PSi/catalyst = 4/1w/w). The mixture was heated at 60 °C for 24 h. Further, the composite fibers were extracted from solution and toluene was removed at 60 °C under vacuum. A 0.12 g/g content of polysiloxane attached to V-MG fibers was calculated by using the Eq. (1):

$$W_{PSi attached} = (W_{Vs-MG-PSi} - W_{Vs-MG}) / W_{Vs-MG}$$
(1)

where: $W_{PSi \ attached}$, $W_{Vs-MG-PS}$, W_{Vs-MG} represent the content of polysiloxanes copolymer attached to the composite fibers (g/g) and the weights (g) of hydropobized and pristine composite samples, respectively.

2.6. Methods

Decane, dodecane and two types of commercial motor oils were used for the sorption experiments. The properties of the testing liquids are given in Table 1.

 Table 1. Properties of oily liquids used in sorption experiments

Liquids	Viscosity [cP]	Density [g/cm³]	Weight loss after 24 h at 30 °C [%]
Decane- C_{10}	0.85	0.72	18 - 22
Dodecane-C ₁₂	1.37	0.74	1 - 2
Motor oil 5W/40	133	0.85	0
Motor oil 15W/40	279	0.88	0

2.6.1. Sorption capacity

The oil sorption capacity was determined according to the known ASTM F 726-99 and CAN/CGSB-183.2-94 standard methods for testing of oil spill sorbents (ASTM, 1999; CAN/CGSB-183.2-94, 1994). Thus, a glass beaker was filled with pure oily liquid (liquid height, 2.5 cm) and a sorbent sample of known weight (around 0.15-0.2 g) was placed into the oil bath and left there for saturation (15 min). After that, the sorbent sample saturated with oil was removed with a magnet (Fig. 1) from the liquid and

left 30 seconds (drainage time) for dripping. Subsequently, it was weighted using a digital balance with 0.001 g resolution. The difference between the loaded (saturated) and pristine sorbent sample gave the maximum sorption capacity (Eq. 2):

$$S = \frac{M_s - M_0}{M_0} \tag{2}$$

where S denotes the sorption capacity, g (oil) / g (sorbent); M_s is the weight of sorbent saturated with oil product (g) at drainage time of 30 s; and M_0 is the weight of pristine sorbent sample (g).



Fig. 1. Recovery of the oil saturated Vs-MG-PSi composite from the oil bath with a magnet

2.6.2. Retention profile

The retention efficiency as a function of time (retention profile) was determined by a gravimetric method. The spent sorbent (loaded with oil) was hanging on a digital balance, whereupon the soaked oil began to drip. The weight of the sorbent was recorded during free drainage of liquid at different time intervals. The retention efficiency was calculated as the ratio of sorption capacity at drainage time (t) over the initial (maximum) sorption capacity (Eq. 3):

$$R = \frac{S(t)}{S(t_{init})} \times 100\%$$
(3)

where *R* is the retention efficiency (in percentage, %); $S(t_{\text{init}})$ is the maximum sorption capacity determined for the initial drainage time $t_{\text{init}} = 0.07 \text{ min}$ (i.e. 4 seconds) and S(t) is the sorption capacity determined for drainage time $t \ge t_{\text{init}}$ (i.e. $t = \{0.07; 0.28; 0.5; 2; 5; 7; 10; 15; 30\}$ min).

2.6.3. Retention capacity after centrifugation

After sorption profile test, the spent sorbent sample was centrifuge at a speed of 2000 rpm for 2 min and weighted. The retention capacity (after centrifugation) was determined by the gravimetric method, representing the ratio of the residual oil weight to dry sorbent weight (Eq. 4):

$$S_{C} = \frac{M_{C} - M_{0}}{M_{0}}$$
(4)

where: S_C is the retention capacity after centrifugation, g (oil) / g (sorbent); M_c is the weight of the sorbent containing the residual oil remained after centrifugation (g); M_0 is the weight of fresh sorbent sample before oil immersion (g). All tests (sorption and retention) were carried out at room temperature (22 ± 2 °C) and the results represent the average values of three trials.

2.7. Characterization

The structures of the Vs, PSi, MG, Vs-MG, Vs-MG-PSi samples were investigated by FTIR spectroscopy on potassium bromide pellets by using a Bruker Vertex 70 Spectrometer. The surface morphologies were visualized by SEM microscopy on ESEM Quanta 200 Electronic Deflection Microscope. For magnetic measurements (magnetization curves) a Quantum Design-PPMSQD-9 vibrating sample magnetometer (-20÷20 kOe range for applied magnetic field) was used. The water contact angles of Vs and Vs-MG-PSi composites were investigated with eScope Conrad USB digital microscope and ImageJ software. The molecular weight and molecular weight distribution of PSi copolymer were measured with a Spectra Physics 8800 gel permeation chromatograph (GPC) equipped with two PL-gel packed columns (103 Å and 500 Å) in THF (tetrahydrofuran) solvent, versus polystyrene standards.

3. Results and discussion

The viscose-based composite characterized by magnetic and hydrophobic properties was prepared by a multistep procedure. First MG composite particles composed of goethite (a-FeOOH) acicular nanoparticles surrounded by spherical maghemite nanoparticles (M) were prepared (Rotaru et al., 2017). A mixture of Vs and MG powder was then submitted to ultrasonication for 15 min, when the temperature increases up to 86 °C to give Vs-MG composite. The final step consisted of the hydrophobization of Vs-MG composite fibers by treating them with a hydrophobic PSi copolymer in the presence of zinc octoate catalyst.

3.1. Structural characterization

The structures of the intermediate and final products were investigated by FTIR spectroscopy (Fig. 2). As one may see from Fig. 2, following ultrasonication of Vs and MG mixture and under increased temperature (up to 86 °C) the bands located at 795 and 885 cm⁻¹, specific to OH groups of goethite in MG spectrum disappeared in the spectrum of Vs-MG composite, while the OH band centered at 3350 cm⁻¹ in the spectrum of Vs-MG composite. Moreover, the C-O-C band of Vs between 1018 and 997 cm⁻¹ is enlarged between 1056 and 1022 cm⁻¹.

All these evidences represent arguments for a strong interaction between MG nanoparticles and Vs. As for the composite fibers treated with polysiloxanes copolymer, the C-O-C specific bands of Vs are practically covered by those of Si-O-Si groups and appear together between 1097-1030 cm⁻¹. The Si-H specific band of PSi copolymer is strongly diminished in the spectrum of Vs-MG-PSi denoting its oxidation and condensation (crosslinking) during ultrasonication treatment (Cojocaru et al., 2017).

The results of scanning electron microscopy (SEM) for the investigated materials are shown in Fig. 3. According to Fig. 3, the diameter of viscose fibers is lower than 10 microns. Vs-MG composite shows agglomerates of maghemite-goethite particles dispersed around the slightly expanded fibers of viscose while for Vs-MG-PSi hydrophobic composite a thin submicronic film of PSi surrounding the viscose fiber can be seen.

3.2. Wettability study

The wettability of Vs and VS-MG-PSi materials was investigated by placing water droplets on the solid surface of fibers and recording the microscopic images (Fig. 4). The images for Vs (Fig. 4, top) indicated that the water droplet was absorbed immediately into the capillary structure of the viscose fibers, and the time for sorption was very short, i.e. less than two seconds. In contrast, the Vs-MG-PSi fibrous sample clearly showed increased hydrophobic property. Its water-contact-angle value was equal to 93 ± 1 degrees. Moreover, when introduced in water, Vs and Vs-MG composite fibers are immediately humidified and migrate to the bottom of the vessel, while Vs-MG-PSi hydrophobic composite fibers stay for indefinite time (the test was accomplished for seven days) at water surface.

3.3. Magnetic properties

The magnetic properties of Vs-MG-PSi hydrophobic composite were determined at room temperature (Fig. 5). The hydrophobic viscose-maghemite-goethite composite synthesized by ultrasonication showed a hysteresis loop specific for magnetic materials. The saturation magnetization value was equal to 26.7 emu/g. From practical standpoint, this composite material loaded with oil can be recovered from the polluted water by applying an external magnetic field (Fig. 1).

As one can see, the pressed viscose material (Vsp) showed the smallest sorption capacity, about 1 g/g for pure hydrocarbons and about 2 g/g for motor oils. The pristine viscose revealed the highest sorption capacities, i.e. 19-20 g/g for decane/dodecane uptake and 41-44 g/g for motor oils uptake. In turn, the hydrophobic composite (Vs-MG-PSi) demonstrated somewhat lower sorption capacities than Vs sample, i.e. 16-17 g/g for decane/dodecane uptake.



Fig. 2. Infrared spectrum of viscose (Vs), maghemite-goethite (MG), poly(hydromethyl-dimethyl)siloxane copolymer (PSi), viscose-maghemite-goethite (Vs-MG) and viscose-maghemite-goethite hydrophobic composites (Vs-MG-PSi)



Fig. 3. SEM micrographs for viscose (Vs), Vs-MG composite and Vs-MG-PSi hydrophobic composite



Fig. 4. Microscopic images recorded at different time intervals showing the wetting of Vs and VSC fibers with water droplets



Fig. 5. Hysteresis loop at room temperature for Vs-MG-PSi composite

However, the hydrophobic magnetic composite shows sorption capacities similar to other reported sorbents or even higher (Table 2), while superior performance of viscose is not of practical interest for oil spill uptake from aqueous media due to its hydrophilic nature.

The retention of oil by the sorbent material is a key factor to assess the ability of the sorbent to retain the oil during transfer and handling operations (Rotaru et al., 2014). In this regard, the retention profile is an important test for the evaluation of the oil drainage from the spent sorbent against time. The results of the retention profile test are shown in Fig. 7 revealing a dramatic drop of the retention capacity of both Vs and Vs-MG-PSi samples (with about 15% for decane and dodecane and about 25% for motor oils) in the time interval ranging from 4 to 30 seconds. For greater values of the draining time (i.e. from 30 seconds to 30 minutes) the retention capacity decreased slowly and attained a steady state at about 86% for hydrocarbons and about 75% for motor oils. These results suggest

that a drainage time of 30 seconds is a proper parameter to estimate the sorption capacity of a material after the removal by dripping of the excess liquid fraction.



Fig. 6. Sorption capacity for Vsp, Vs and Vs-MG-PSi sorbent materials

The results of the centrifugation test are shown in Fig. 8. Under the centrifugal force, 76-77 % of decane and dodecane liquids can be recovered from Vsp and 98-99% from Vs and Vs-MG-PSi spent sorbents. In turn, the motor oil can be recovered in a percentage of 85-86% from Vsp, and 94-95% from Vs and Vs-MG-PSi loaded materials.

4. Conclusions

In this work a hydrophobic viscosemaghemite-goethite composite with magnetic properties was produced as a potential sorbent for oil spill cleanup from aquatic media. The composite was characterized by FTIR, SEM, hydrophobicity and magnetic measurements. No wettability was observed by maintaining the composite on water surface for long period of time. The saturation magnetization of the developed composite showed a value of 26.7 emu/g, appropriate for facilitating the magnetic separation of the spent sorbent (loaded with oil).

Material (sorbent)	Oil type	Sorption (g/g)	Form	Reference
Peat	Diesel oil	9-12	Granular	Cojocaru et al. (2011)
Clay-polymer aerogel	Dodecane	23.6	Aerogel	Rotaru et al. (2014)
Hydrophobized clay-polymer aerogel	Dodecane	10.5	Aerogel	Rotaru et al. (2014)
Sawdust	Heavy crude oil	4.1-6.4	Granular	Annunciado et al. (2005)
Polyurethane	Diesel oil	40-50	Foam	Li et al. (2012)
Hydrophobized polyester fibers	Motor oil	10.03	Fibers	Cojocaru et al. (2017)
Pristine viscose	Decane/Dodecane	19/20	Fibers	This work
	Motor oil	41/44	Fibers	This work
Vs-MG-PSi composite	Decane/Dodecane	16/17	Fibers	This work
	Motor oil	36/38	Fibers	This work

Table 2. Oil sorption capacity of Vs-MG-PSi composite as compared to those of pristine viscose and other reported sorbents







Fig. 8. Retention capacity (Sc) after centrifugation of the spent sorbents VsP, Vs and Vs-MG-PSi

The results of sorption and retention tests following revealed the order of sorption performances: Vsp << Vs-MG-PSi < Vs. Although the sorption capacity of the hydrophobic magnetic composite is slightly lower than that of the pristine viscose, it is similar or even higher as compared to other reported sorbents. Thus, the Vs-MG-PSi hydrophobized magnetic composite can be use as oil sorbent in aquatic media. Moreover, the composite has the advantage of an easy magnetic recovering after sorption.

Acknowledgments

This work was supported by a grant of the Romanian National Authority for Scientific Research and Innovation, CNCS – UEFISCDI, project number PN-II-RU-TE-2014-4-1266.

References

- Abdul A., Adebayo A.R., Hossain M.E., (2012), A novel sustainable oil spill control technology, *Environmental Engineering and Management Journal*, 13, 334-353.
- Abdullah M.A., Rahmah A.U., Man Z., (2010), Physicochemical and sorption characteristics of Malaysian Ceiba pentandra (L.) Gaertn as a natural oil sorbent, *Journal of Hazardous Materials*, 177, 683-691.
- Annunciado T.R., Sydenstricker T.H.D., Amico S.C., (2005), Experimental investigation of various vegetable fibers as sorbent materials for oil spills, *Marine Pollution Bulletin*, **50**, 1340-1346.
- ASTM F 726-99, (1999), American Society for Testing and Materials, Standard Method for Testing Sorbent Performance of Adsorbents, USA & Canada.
- CAN/CGSB-183.2-94, (1994), *Method for Testing Sorbents*, Canadian General Standard Board, Canada.
- Cojocaru C., Macoveanu M., Cretescu I., (2011), Peat-based sorbents for the removal of oil spills from water surface: Application of artificial neural network modeling, *Colloids and Surfaces A*, 384, 675-684.
- Cojocaru C., Pricop L., Samoila P., Rotaru R., Harabagiu V., (2017), Surface hydrophobization of polyester fibers with poly(methylhydro-dimethyl)siloxane copolymers: experimental design for testing of nonwoven materials as spill sorbents, *Polymer Testing*, **59**, 377-389.
- Husseien M., Amer A.A., El-Maghraby A., (2008), Experimental investigation of thermal modification influence on sorption qualities of barley straw, *Journal* of Applied Sciences Research, 4, 652-657.

- Johnson R.F., Manjrekar T.G., (1973), Removal of oil from water surfaces by sorption on unstructured fibers, *Environmental Science and Technology*, 7, 439–443.
- Khan E., Virojnagud W., Ratpukdi T., (2004), Use of biomass sorbents for oil removal from gas station runoff, *Chemosphere*, **57**, 681-689.
- Likon M., Remškar M., Ducman V., Švegl F., (2013), Populus seed fibers as a natural source for production of oil super absorbents, *Journal of Environmental Management*, **114**, 158–167.
- Li H., Liu L., Yang F., (2012), Hydrophobic modification of polyurethane foam for oil spill cleanup, *Marine Pollution Bulletin*, **64**, 1648-1653.
- Pricop L., Hamciuc V., Marcu M., Ioanid A., Alazaroaie S., (2005), Graft copolymers polydimethylsiloxanepolyethyleneoxide. Synthesis and characterization, *High Performance Polymers Journal*, **17**, 303-312.
- Radetic M., Ilic V., Radojevic D., Miladinovic R., Jocic D., Jovancic P., (2008), Efficiency of recycled wool-based nonwoven material for the removal of oils from water, *Chemosphere*, **70**, 525-530.
- Ribeiro T.H., Rubio J., Smith R.W., (2003), A dried hydrophobic aquaphyte as an oil filter for oil/water emulsions, *Spill Science & Technology Bulletin Journal Impact B*, **8**, 483-489.
- Rotaru A., Cojocaru C., Cretescu I., Pinteala M., Timpu D., Sacarescu L., Harabagiu V., (2014), Performances of clay aerogel polymer composites for oil spill sorption:

Experimental design and modeling, *Separation and Purification Technology*, **133**, 260-275.

- Rotaru R., Samoila P., Lupu N., Grigoras M., Harabagiu V., (2017), Maghemite (γ-Fe₂O₃)/goethite (α-FeOOH) ferromagnetic composites obtained through ultrasonication, *Revue Romaine de Chimie*, **62**, 131-138
- Senanurakwarkul C., Khongsricharoen P., Pejprom D., Tantayanon S., Khaodhiar S., (2013), Effects of the composition and the preparation methods on oil sorption capacity of recycled rayon waste-kapok mixtures (RRWK) sorbent, *International Journal of Environment and Sustainable Development*, 4, 246-250.
- Srinivasan A., Viraraghavan T., (2010), Oil removal from water using biomaterials, *Bioresource Technology*, 101, 6594-6600.
- Suni S., Kosunen A., Hautala M., Pasila A., Romantschuk M., (2004), Use of a by-product of peat excavation, cotton grass fibre, as a sorbent for oil-spills, *Marine Pollution Bulletin*, **49**, 916-921.
- Vazquez F., (1999), Silicones: Beyond Softening in Garment Finishing, Book of papers, AATCC Garment Finishing Symposium.
- Weber W.J.J., McGinley P.M., Katz L.E., (1991), Sorption phenomena in subsurface systems: concepts, models and effects on contaminant fate and transport, *Water Research*, 25, 499-528.