Application of Hollow Fibre Membrane Bioreactor Instead of Granular Activated Carbon Filtration for Treatment of Wastewater from Car Dismantler Activity

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Nowadays, membrane technologies are becoming more frequently used for separation of wide varying mixtures result from a lot of industries and can compete successfully with traditional schemes. The aim of this paper was to analyse the use of ultrafiltration with polypropylene hollow fibre membrane in the chemical physical systems instead of Granular Activated Carbon (GAC) filtration. A process data collection was performed and integrated with a characterization of the process effluents in terms of treatability and reusability. In order to evaluate properly the wastewater loading, an analysis course was set. The samples have been gathered for two years; instantaneous samples were drawn from the influent and treated wastewater. Based on daily average values, a general average has obtained. The experimental data were statistically analysed and the average values of the investigated parameters. It was found that ultrafiltration is a good process solution before discharge of the effluent.

Keywords: hollow fibre, membrane MBR, GAC, filtration, wastewater treatment

Water is essential for life, and although covering approximately 70% of the earthly crust area, only a small fraction of the water is actually compatible with terrestrial life forms [1]. Water scarcity in many parts of the world is a significant concern [2]. With increasing of global population, the gap between the water supply and demand is broadening and is reaching such alarming levels that in some parts of the world it threatening human existence [3].

Water scarcities across many countries have meant that recycling and reuse of wastewater has become a priority. With the improvement of water/wastewater treatment techniques, a suite of technologies is now available to provide a higher standard and to allow better end-use application of the treated effluents in an urban environment [4,5]. These include technologies such as sand filtration, adsorption using granular activated carbon, zeolite or other clay materials, membrane MBR bioreactor, which can be applied based on the treated wastewater quality requirements and matching specific end-uses [4].

Automobile recyclers and dismantlers are in the business of taking otherwise discarded products and reusing or recycling them. The discarded cars and waste parts can feature a high environmental impact if proper precautions are not taken [4-6]. Vehicle salvage businesses generate process wastewater during daily operations from equipment cleaning, car washing, paint spray booths or other sources [5].

Wastewater from auto recycling activities may contain contaminants, such as gasoline or diesel fuel, transmission and brake fluids, solvents, hydrocarbons in general, grease, oil, dirt, detergents and chemical residue from cleaners such as heavy metals of various kinds, scrap metal, (e.g mercury from switches and mercury-containing lamps, lead from lead-acid batteries), chlorofluorocarbons (CFCs) and other refrigerants from air conditioning units, waste tires and other contaminants [7].

Proper management and disposal of wastewater is essential to protect public health and the environment. If not managed properly, wastewater and its contaminants can negatively impact surface waters and groundwater. Two main types of wastewater are typically generated by auto recycling facilities: *Sanitary wastewater* includes wastewater generated from normal use of lavatories, washrooms, showers, drinking fountains, etc (Sanitary wastewater can be discharged to a city sewer system or a DEQ approved onsite wastewater treatment (septic) system) and *Process wastewater* includes wastewater going into floor drains in dismantling and work areas: aqueous cleaning, steam cleaning, equipment wash down water, or water from other sources where it comes into contact with dismantled parts or equipment [3,6,8].

All industrial wastewater should go to an on-site treatment plant, and should never discharge to the ground, storm water system, septic system, or drywell [8, 9].

Recent soil and water protection regulations require wastewater from storage yard wash outs due to rain, to be treated before being drained into groundwater bodies. In particular, all of the wastewater that is generated every time it rains must be treated, since the wash out of the piles of scrap and their respective storage areas, with their oil load, lasts for the entire duration of precipitation [5-9].

The car demolition activity it is certainly presented as a high environmental risk activity with not only for the inherent in objective risks inside which there are significant quantities of dangerous substances (acids, mineral oils, fuels, etc.), but also for the presence of combustible materials and flammable substances, noisy activities, activities and storage places outside pollutants [10,11].

The aim of this paper was to analyse the use of ultrafiltration in the chemical physical systems instead of GAC filtration coal sources of water pollution and loading concentrations in car dismantlers sector.

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Experimental part

Materials and methods

Study area

The present study was conducted at Autodemolizioni Modulor, Farra d'Isonzo Gorizia - a car dismantler company in Italy.

Fig.1. The Autodemolizioni Modulor company area

Full-scale design, setup and description

The real scale plant was built by GOST Ltd and run a chemical- physical treatment process composed of pretreatment (primary sedimentation), oil removal, coagulation, neutralisation, flocculation, and initially, rapid sand and GAC filtration.

The upper settling tank of the chemical-physical treatment system had both accumulation and separation (from settleable/floating materials) function.

The wastewater, subsequent to separation of the solid matters, flows through a proper flow regulator in the oil removal tank. The separation and recovery of the oil does not require the presence of other external tanks because a special container for its gradually collecting is provided.

The chemical treatment process takes place within dedicated four tanks where are dosed the specifically selected reagents indispensable to the treatment process. Each tank is equipped with air stirring complete of flow regulator.

The treated water output from chemical treatment tanks, flows into the sedimentation tank where the separation from the sludge takes place and, once clarified, is entered the filtration/adsorption system: a pump aspires it and pushes it through the filters, first quartz sand and then granular activated carbon (GAC).

The quartz sand filter is provided with backwashing and clogged filter indicator. The purification system is completed with a tank for pick up the first rain waters which are then processed according to the imposed standards.

Real wastewater was collected from the activities process and contains suspended solids (TSS), suspended and dissolved organics (COD, BOD_5), grease and oils, transmission and brake fluids, solvents, hydrocarbons (gasoline or diesel fuel), heavy metals (e.g mercury, lead), waste tires and other contaminants.

Wastewater was collected once a month and kept in the freezer for further use. The characteristics of the wastewater samples are listed in Table 1. The pH can vary between 4.5 and 6.5, TSS between 600 mg/L and 2000 mg/L, FOG between 10 and 2000 mg/L, COD between 3000 and 35000 mg/L.

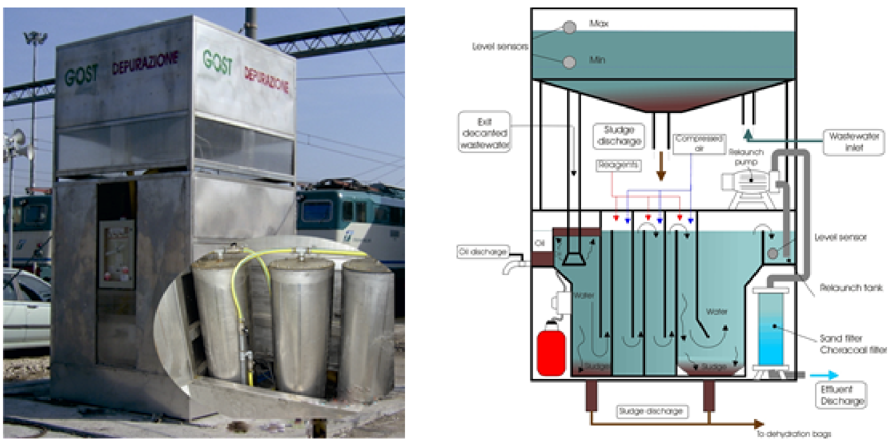
The GAC used

The coal based granular activated carbon GAC of mineral origin physically activated (UNIVARCARB GAC 830 from Univar Italy) was used in this study. This coal based GAC has the subsequent specifications: Iodine number (ASTM D 4607) min. 800 mg/g, apparent density (AWWA B 600-78) 480 ± 40 kg/m^3, specific surface area (B.E.T.) min 800 m^2/g, moisture packaging (ASTM D 2867) 5% max, ash (ASTM D 2866) max 12%, pH (ASTM D 3838) basic, granulometry (ASTM D 2862)> 8 mesh max 5%, <30 mesh max 5%, typical contact time of 8-30 min, bed height 1-3 m, linear velocity 5-20 m / h, bed expansion to the backwash ca 20%.

There are three GAC filters, but they are efficient only for a short time due to quick exhaustion which might result in frequent backwashing and unsatisfactory filtration efficiency and must be often regenerated. However, the dual use of GAC as both adsorbent and filter medium for solids retention in advanced wastewater treatment is a novel concept and has not been investigated sufficiently up to now.

While GAC has previously been applied for adsorptive removal of organic pollutants from wastewater [11], high COD (chemical oxygen demand) concentrations are usually seen as unfavourable for adsorption onto GAC, because of long-term carbon fouling reducing the adsorption capacity and leading to accelerated breakthrough [12-14].

Adsorption is both the physical and chemical process of accumulating a substance at the interface between liquid and solids phases. Adsorption is a reversible process. As some molecules attach themselves to the surface, others are kicked off back into the fluid. At equilibrium, the

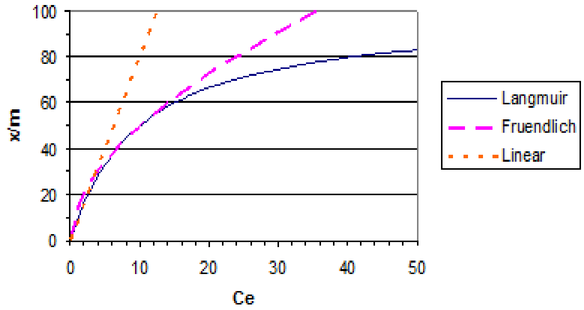


a *b*

Fig. 2. The flow scheme of chemico-physical plant with sand and coal filters (a-general view; b-flow scheme)

Table 1
CHARACTERISTICS OF WASTEWATER

Parameter	pH	TSS	COD	Oil & grease
Average (mg/L)	4.5 – 6.5	600 – 2000	1.500 – 10.000	10 - 20000



Langmuir	k	0.1
	$(x/m)_{max}$	100
Freundlich	K	14
	1/n	0.55
Linear	K_p	8

Fig. 3. Adsorbtion isoterms

molecules movement rate *onto* the surface (adsorption) and *off* the surface (desorption) are equal. Both rates are concentration dependent.

In order to examine the relationship between adsorbate concentrations in the solid and aqueous phases at equilibrium, sorption isotherm models are widely employed.

The Langmuir and Freundlich models are most widely used to model the equilibrium data of adsorption [15-19].

Langmuir isotherm is a theoretical equation:

$$\frac{x}{m} = \frac{k(x/m)_{max}\, C_e}{1 + kC_e} \qquad (1)$$

where: k is an empirical coefficient and $(x/m)_{max}$ is the amount of adsorbate that would form a monomolecular layer on the solid adsorbent. In essence, it is the maximum mass of adsorbate that could be retained.

Freundlich developed the following empirical isotherm:

$$\frac{x}{m} = KC_e^{\,1/n} \qquad (2)$$

where: K and 1/n are empirical constants.

A third isotherm which is commonly used in groundwater studies is the Linear Partitioning isotherm:

$$\frac{x}{m} = K_p C_e \qquad (3)$$

where: K_p is an empirical coefficient. Although the three equations look different, they can be thought of as curve fits to the same phenomenon. In the graph below, all three isotherms are plotted together. The coefficients used are listed in the table below.

After a long time process monitoring with not always good results in order to obtain an effluent that meets the standards for discharging in watercourses, two years ago, the GAC filters were substituted with a polypropylene hollow fibre MBR module and the effluent was monitored for 12 months.

Membrane characteristics

The membrane ability depends on the pores size, type of material, wastewater type which has to be treated, solubility and retention time [20, 21].

GOST MBR membrane filtration modules consist of bundles of hollow fibres mounted on a strong AISI 304 stainless steel supporting frame and connected by Akulon pipes, very resistant and non-deformable material. The manifolds are interlocking inserted into the frame and are connected to the suction tube with PVC pipes with quick mounting. The support structure of the modules is used to give rigidity to the system. The height of the frame is sufficient to ensure that the fibres remain rather *soft*, or better, able to move under air action blown from below the module (fig.4, table 2).

To further reduce any mud, the modules have an integrated air distribution system under the fibres through a blower. The air flows as bubbles along the fibres generating a higher turbulence and minimizing the biomass storage on the fibres themselves. Also the system allows a greater degradation of refractory organic compounds [22]. Indeed, the high molecular weight that often characterizes these compounds makes waterproof membrane and therefore significantly increases the contact time in the activated sludge tank, favouring the specific bacteria consortia development [23]. The hollow fibres are PP superficially modified to ensure optimal porosity, able to remove all suspended solids, colloids, bacteria and cysts [24-28].

The solid-liquid separation occurred in the MBR tank equipped with 1 module submerged hollow fibre membrane with a nominal pore size of 0.02 – 0.002 ¼m (GOST Ltd.). The total membrane area was 126 m^2. The membrane was operated with an on/off cycle aimed to provide a relaxation time in such way that every 2 min the permeate discharge was stopped about 15 s for cleaning

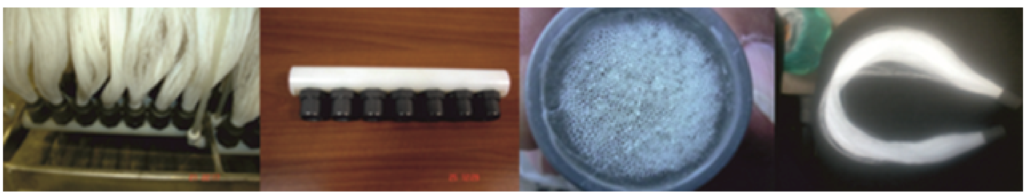


Fig.4. GOST submerged hollow fibre membrane module and bundle (courtesy of GOST MBR solutions)

Table 2

MAIN MEMBRANES FEATURES

Fibres material	Polypropylene
Porosity	40 – 50%
Pore size	0,02 -0,2 µm
Outer fibre diameter	0,45 mm
Washing conditions (pH)	7
Washing conditions (temperature)	Tmax = 50 °C
Backwash	SI
Bundle size	Φ 25 x 750 mm
Bundle	1000 fibre
Filtration area of a bundle	1,00 m^2
Filtration surface of a module	126 m^2
Working pressure	0,1 – 0,4 bar
Permeate flow average	10 – 15 l/ m^2 h

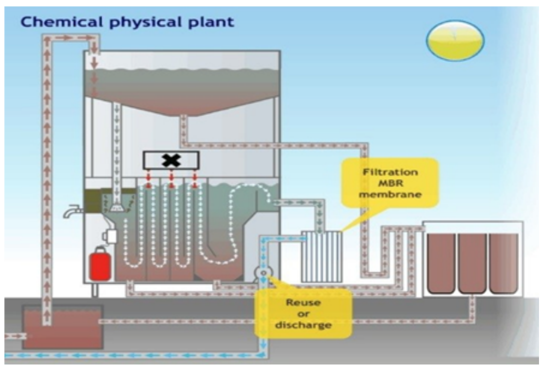


Fig. 5. Chemico- physical plant with MBR

through backwashing, and the daily operating time of membrane was 21 h. Membrane fouling was reduced by introducing air at the bottom of the membrane module (scouring) as well as by the on-line backwashing with tap water [26].

The sludge was discharged from MBR daily to maintain a relatively stable MLSS concentration and a desired solids retention time (SRT).

Membrane bioreactor processes have been widely used to reduce or eliminate organic pollutants, heavy metals and to provide a superior rating for most bulk water quality indicators [27-39].

Table 3 shows the characteristics of a hollow fibre. The sample was conditioned in standard conditioning atmosphere according to EN ISO 139/2005.

Figure 6 shows images of polypropylene hollow fiber before filtration.

Sample collection

Samples were collected in plastic bottles from the effluent channel and transferred to the laboratory, preserved and stored at 4°C for further analytical determinations and treatment. Following collection, samples were stored until their utilization.

The influent and effluent samples were collected regularly, one time per month, to investigate the GAC and MBR filtration performance. The samples of the effluent were characterized for determining *p*H, total alkalinity, chemical oxygen demand and dissolved chemical oxygen demand (COD), chloride, total nitrogen, total suspended solids, Al, Cu, Pb, SO_4, ammonia nitrogen (NH_4^+-N), nitrate nitrogen (NO_3^--N), total surfactants (Tt). All analyses were performed according to Standard Methods for the Examination of the recommendations of Water and Wastewater [27-29,38].

Physicochemical parameters

In order to measure the above parameters were used photochemical commercial test kits (Hach Lange GmbH, Düsseldorf, Germany) LCK type. The pH measurements were done using digital pH meter (Hanna Instruments, Italy). The spectrophotometric analysis was done using XION 500 Dr Lange spectrophotometer (Hach Lange, Italy). Concentrations of total suspended solids (TSS) were determined gravimetrically after filtration of 2.5 L influent sample and up to 10 L effluent sample through glass fibre filters (0.3 e 1.0 mm, MachereyeNagel, Germany). Quantification of dissolved parameters was preceded by

Table 3
PHYSICAL CHARACTERISTICS OF MBR FIBRE

Determined characteristics	U.M.	Fibre
Density of length	dtex (den)	198,3(178,5)
	Cv%	6,36
Breaking strength	N	5,63
	Cv%	6,0
Elongation at break	%	69,6
	Cv%	18,48
Breaking strength to knot	N	6,02
	Cv%	7,94
Extension to break to knot	%	79,2
	Cv%	20,34
Breaking strength in wet state	N	5,92
	Cv%	7,34
Elongation at break in wet state	%	66,8
	Cv%	26,17
Breaking strength to knot in wet state	N	5,83
	Cv%	6,49
Extension to break in wet state	%	71,0
	Cv%	27,57
Apparent diameter	µm	445,4
Fibre length	mm	763

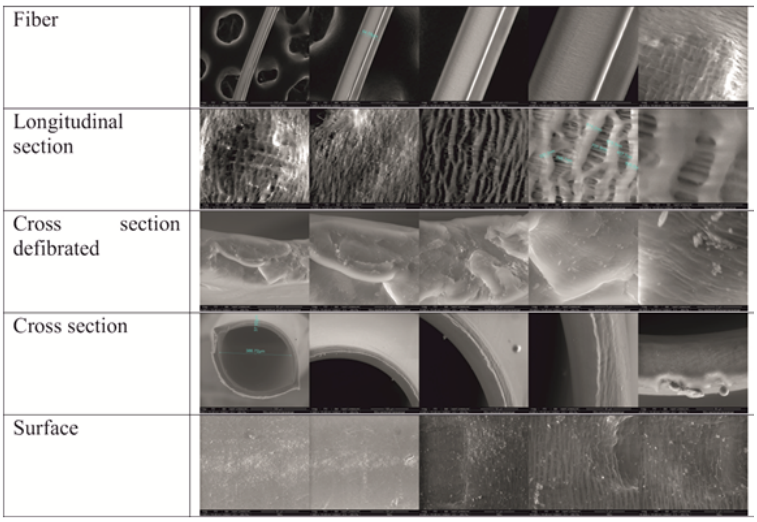


Fig. 6. SEM Images of fiber before filtration

sample filtration using 0.45 mm membrane filters (regenerated cellulose, MachereyeNagel, Germany).

Results and discussions

In this study were performed two experiments: 1st study aimed monitoring the pollutants removal using GAC filtration for two years and 2nd study aimed monitoring the effluent quality using ultrafiltration (UF) with polypropylene hollow fibre MBR membrane instead of GAC filtration for one year. It was found that the filtration efficiency of parameters removal is high. After treatment, the effluent is suitable for reuse, marked by parameters corresponding to the prescribed by regulations in force.

Figure 7 shows the average of the pH, based on samples of the effluent in 1st and 2nd experiments. Furthermore, it was found that variations occurred during the experiment in each phase.

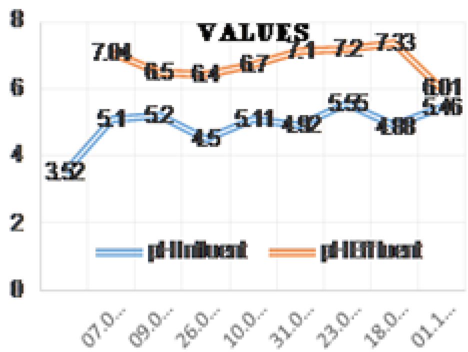


Fig.7. The average of the pH on time

COD removal efficiency

Results for COD removal efficiency in the treatment plant during the experiment period in the influent and final effluent during the 1st and 2nd phase treatment are presented in figure 8. The results indicated that the COD concentrations in the effluent for the 2nd phase were all lower than the ones from the 1st phase. Despite the wide range of COD concentration in the influent, from 1500 to 10000 mg/L, with an average value of 4750 mg/L, the combined system showed significant performance in organic carbon removal in particularly in the 2nd phase when was used filtration with MBR. The COD removal efficiency in the 1st phase

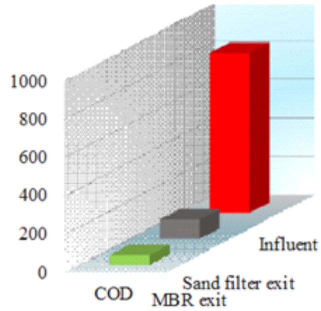


Fig. 8. The COD removal evolution

was in the range 70–80%. The removal efficiency increased in the 2nd phase, when additional ultrafiltration membrane MBR entailed a reduction of the studied parameters values by an additional 10 to 15%. It can be seen that the average efficiency of COD removal is approximately 90% compared to only 75% that it notes in 1st phase. In the 2nd phase was installed 1 MBR module and were eliminated the three GAC filters. The results obtained for removal efficiency are showed in figure 8 and membrane aspects in figure 9.

In the figure 9 are presented the SEM images after one-year functioning.

From the SEM was observed that inside the hollow fibres tubes, a residue was formed. The residue characterization is pesented in table 4 and figure 10.

Heavy metals removal

Heavy metal pollution is among one of the most concerning environmental problems in our days. For this reason, the treatment of wastewaters containg heavy metals is a very important issue due to their recalcitrante and persistence in the enviroment [30-40]. Heavy metals are elements having atomic weights between 63.5 and 200.6, and a specific gravity greater than 5.0 [30-37].

Chemical precipitation is effective and by far the most widely used process in industry [31] because it is relatively simple and inexpensive to operate. In precipitation processes, chemicals react with heavy metal ions in order to form insoluble precipitates. The precipitates formed can be separated from the water by sedimentation or filtration. It was used hydroxide precipitation due to its relative simplicity, low cost and ease *p*H control [32, 36]. The solubility of the various metal hydroxides is minimized in

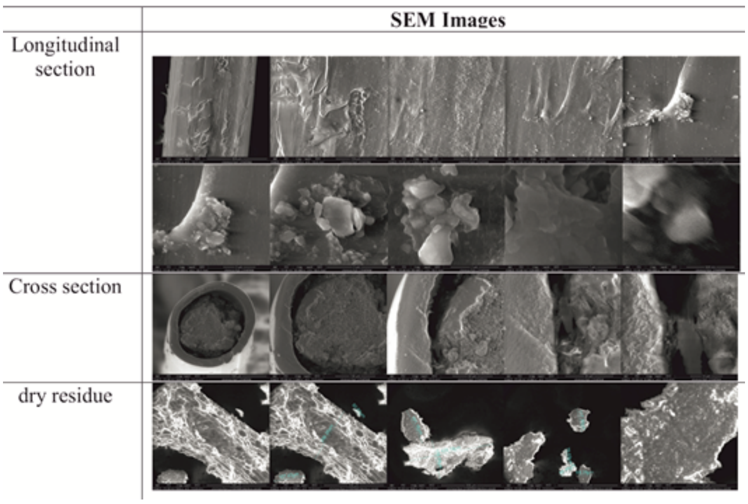


Fig. 9. SEM images of fibre after one year

Table 4
DRY RESIDUE

Wt(dry residue)	C	O	Na	Mg	Al	Si	P	S	K	Ca	Fe
%	49.80	28.52	0.91	0.32	2.34	3.36	3.89	1.63	0.41	6.85	1.97

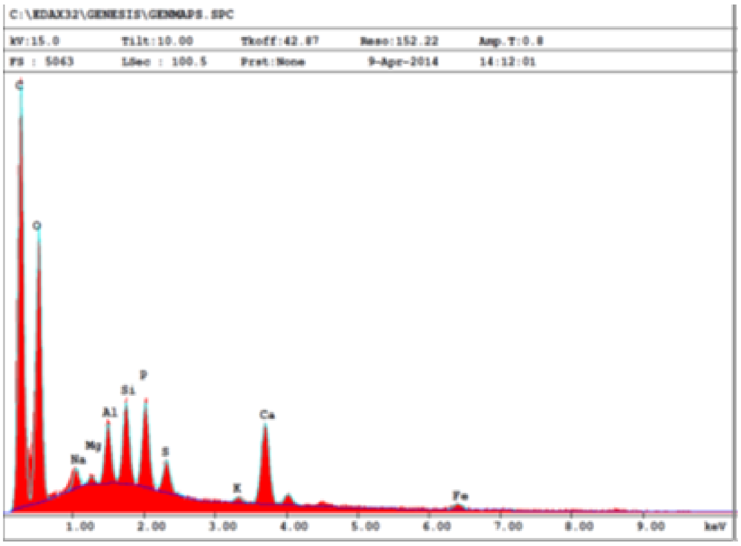


Fig. 10. EDAX Spectrum for dry residue

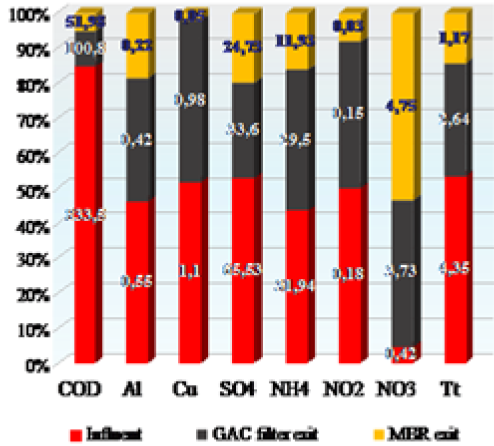


Fig. 11. Removal efficiency for studied parameters

Table 5

REMOVAL EFFICIENCY FOR STUDIED PARAMETERS

		COD	pH	Al	Cu	SO_4	NH_4	NO_3	NO_2	St
Influent	mg/L	833,50	6.96	0.55	1.10	65.53	31.94	0.42	0.18	4.35
Effluent GAC	mg/L	100.80	5.76	0.42	0.23	33.60	29.05	3.73	0.15	2.64
Effluent MBR	mg/L	51.98	7.06	0.22	0.05	24.73	11.93	4.75	0.03	1.17

the *p*H range of 8.0 - 11.0. The metal hydroxides were removed by ûocculation and sedimentation [33, 40].

The removal results of task chemical species from this study through membrane were presented in table 5 and figure 11.

Conclusions

The adsorption process on GAC is reversible and difficult to control. Depending of wastewater characteristics, the GAC saturation occurs more or less rapidly.

The UF in most of the cases is a good solution because:
- the effluent is micro sludge-free and can be reused;
- ensures a further reduction of the parameters;
- eliminates the high costs of management and GAC disposal and/or regeneration.

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References

1.SHIKLOMANOV, I., chapter World fresh water resources, Peter H. Gleick, Water in Crisis: A Guide to the World's Fresh Water Resources, Ed.Oxford University Press, New York, 1993

2.ELIMELECH, M., PHILLIP, W.A., Science, **333**, 2011, p.712-717

3.ALOIS, P., Arlington Institute, 2007

4.HO, J., SUNG, S., Wat.Res., **44**, 2010, p.1409-1418

5.*** Auto Dismantler Handbook, Best management practices and environmental compliance, 2015

6.PATAKI, G. E., CROTTY, E. M., Environmental Compliance and Pollution Prevention Guide for Automobile Recyclers, 2003

7.PATAKI, G. E., CROTTY, E. M., Environmental Compliance and Pollution Prevention Guide for Small Quantity Generators, A summary of regulations, for air, water and hazardous waste. New York State Department of Environmental Conservation, Pollution Prevention Unit, 1998, p.41

8.*** Auto Recyclers Guide to a Cleaner Environment - Best Management Practices Monroe County Small Business Pollution Prevention Task Group and New York State Department of Environmental Conservation, **2001**, p.36

9.GIGLIO, G. A., Guida alle migliori pratiche e tecnologie disponibili nel settore dell'autodemolizione, European Regional Environmental Services Platform

10.MAZUMDER, D., MUKHERJEE, S., Intern.J.Environm.Sci.Developm., **2**, 2011, p.64-69

11.ALBU, P.,C., AL ANI, H. N. A., CIMBRU, A., M., Popa, G. A., Niculae, A. G., Miron A. R., Rev. Chim. (Bucharest), **67**, no. 4, 2016, p. 813

12.CHAUDHARY, D. S., VIGNESWARAN S., JEGATHEESAN, V., NGO, H. H., Moon, H., KIM, S.H., Wat. Sci. Tech., **47**, 2003, p.113-120

13.RIZZO, L., FIORENTINO, A., GRASSI, M., ATTANASIO, D., GUIDA, M., J.Environm.Chem.Eng., **3**, 2015, p.122-128

14.SKOUTERISA, G., SAROJA, D., MELIDISB, P., HAIC, F. I., OUKIA, S., Biores.Techn., **185**, 2015, p.399-410

15.RIKABI, A. A. K. K., CUCIUREANU, A., CHELU, M., MIRON, A. R., ORBECI, C., POPA, A. G., CRACIUN, M. E., Rev. Chim.(Bucharest), **66**, no. 8, 2015, p.1093

16.PARK, S. J., KIM, K., D., Carbon, **39**, 2001, p.1741-1746

17.***https://iaspub.epa.gov/tdb/pages/treatment/treatmentOverview.do?treatmentProcessId=979193564

18.LE CLECH, P., JEFFERSON, B., JUDD, S. J., J. Membr. Sci., **218**, 2003, p.117-129

19.DEFRANCE, L., JAFFRIN, M.Y., GUPTA, B., PAULLIER, P., Biores.Technol., **73**, 2000, p.105-112

20.STEFAN, H., WALTER, T., J. Biotechnol., **92**, 2001, p.95-101

21.CHANG, I.S., LEE, C.H., AHN, K.H., Sep. Sci. Technol., **34**, 1999, p.1743-1758

22.LEE, J., AHN, W.Y., Water Res. **35**, 2001, p.2435–2445

23.GANDER, M., JEFFERSON, B., JUDD, S., Sep. Purif. Technol., **18**, 2000, p.119-130

24.FAZAL, S., ZHANG, B., ZHONG, Z., GAO, L., CHEN, X., J.Environm.Prot., **6**, 2015, p.584-598
25.*** http://www.thembrsite.com/, accesat in 2014
26.***APAT, IRSA – CNR, Metodi analitici per le acque, ISBN 88-448-0083-7, 2004
27.*** American Public Health Association (APHA), American Water Works Association (AWWA), Water Environment Federation (WEF): Standard methods for the examination of water and wastewater. 22. Ed., Washington ,2012
28.SRIVASTAVA, N.K., MAJUMDER, C.B., J. Hazard. Mater., 151, 2008, p.1-8
29.FU, F., WANG, Q., J.Environm.Man., **92**, 2011, p.407-418
30.WANG, J., CHEN, C., Biotechnol. Adv., 24, 2006, p.427-451
31.KU, Y., JUNG, I.L., Water Res., **35**, 2001, p.135-142
32.GAITHER, M., Automotive Mercury Switch Removal Programs-Final Report, Sqg-mercury-2, **8**, 2004
33.ENJARLIS, E., APCBEE Procedia, **9**, 2014, p.145-150
34.PASCU, D. E., MIRON, A. R., TOTU, M., NECHIFOR, A. C., EFTIMIE TOTU, E., JOAM, **17**, 2015, p.1161-1167
35.PASCU, D. E., MODROGAN C., MIRON, A. R., ALBU, P.C., CLEJ D.D., CAPRARESCU, S., Rev. Chim.(Bucharest), **66**, no. 12, 2015, p. 1950
36.NECHIFOR, A. C., NAFTANAILA, L., RIKABI, A. A. K., DINU, A., PANAIT, V., MIRON, A. R., Rev.Chim.(Bucharest), **65**, no. 4, 2014, p.386
37.TANCZOS, S.-K., CHICAN, I., MIRON, A. R., RADU, D. A., RADUCU, A., NECHIFOR, A. C., Rev. Chim.(Bucharest), **65**, no. 6, 2014, p.636
38.MIRON, A. R., NECHIFOR, A. C., RIKABI, A. A. K. K., TANCZOS, S.-K., EEMJ, 14, 2015, p.373-379
39.MIRON, A.R., CHIVU, A.M. A., RIKABI, A. A. K. K., ALBU P. C, Rev. Chim. (Bucharest), **65**, no. 12, 2014, p.1399
40.MIRON, A.R., RIKABI, A. A. K. K., NICULAE A.G.,TANCZOS, S.-K., Rev. Chim.(Bucharest), **65**, no. 1, 2015, p.6

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