Bulgarian Chemical Communications, Volume 48, Special Issue D (pp. 102 – 111) 2016

Investigating the efficiency of nanocomposite membranes synthesized by polyacrylonitrile polymers containing single-walled carbon nanotubes in decreasing chemical and biological pollution indicators of greywater

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Received June 18, 2016; Revised September 10, 2016

One of the most important concerns of human society is successive droughts and water resource shortage. To overcome water shortage crisis, water consumption pattern together with wastewater treatment regarding water resources recycling are necessary. The best and cheapest practical solutions are to decrease per capita water consumption in homes as well as treatment of greywater. The purpose of the present study was to determine the efficiency of nanocomposite membranes synthesized by polyacrylonitrile polymers containing Single-Walled Carbon Nanotubes (SWCN) in decreasing chemical and biological pollution indicators of greywater. To synthesize the membrane of SWCN, polyacrylonitrile polymer with the molecular weight of $1.5 * 10^6$ g/mol and SWCNT produced by US Research Nanomaterials with the purity of 96% and approximate diameters of 9-15nm as well as DMF solvent were used. In order to prepare nanofiber, the electrospinning process with ultrasonic was used. Then, to evaluate the efficiency of the prepared membranes in greywater treatment, parameters such as COD, BOD5, TSS, TDS, Detergent, and Fecal Coliform and Total Coliform were used. Data were analyzed in SPSS 19 and the results were presented using graphs, figures, tables, mean, and standard deviation. Among synthesized nanocomposite membranes, PAN+2.5%CNT showed the highest efficiency. The application of this membrane in treating the gray water led to decreased parameters including COD, BOD5, TDS, Detergent, Fecal Coliform, and Total Coliform by 98%, 89.62%, 91.4%, 88.4%, 90.9%, 100%, and 99.28%, respectively. The results showed that nanocomposite membranes synthesized by polyacrylonitrile polymer containing carbon nanotubes can be effective in decreasing pollution indicators of the greywater with high efficiency. The results showed that the use of nanocomposite membranes containing CNT in greywater treatment can be effective in dealing with water shortage and preserving water resources and environment.

Keywords: Nano-composite, Nano fiber, single-walled carbon nanotubes (SWCNT), gray water, purification, electro spinning, bathroom shower.

INTRODUCTION

Global warming and water shortage are among the most important concerns of human societies in the decades ahead [1]. To deal with these crises, various solutions should be used. One of the practical solutions to preserve water resources is greywater reuse [1,2]. greywater is referred to wastewaters resulted from laundry, shower, and kitchen except toilet [3-5].Of course, many scholars do not consider wastewaters from kitchen among greywater [6-9] .Up to now, numerous efforts have been done regarding greywater treatment and reusing it. One of the most important methods is filtration with soil, using wetlands, rotating biological contactor (RBC), sequenced bath reactor (SBR) reactor, ultrafiltration UF, and membrane bioreactors MBR [10-13]. Parameters including TSS, TDS, BOD, COD, pathogens, coliforms, detergents, nitrogen compounds, and phosphates are among the most important pollution factors o of greywater. The results of a study showed that with greywater reuse in toilet's flash tank, about 30% water consumption saving occurs[14] Also, it is possible to use this water to wash car, irrigation of golf courses, camps [15], fire-fighting, boilers, or cement production [16]

Various studies have shown that the use of Singe Wall Carbon Nanotubes (SWCNTs) can increase the flow passing through the polymer membranes. The results of a study showed increasing the flow by 50000 time in carbon nanotubes led to attention to producing carbon nanotubes in industrial scale [17]. Polymers, particularly polyacrylonitrile (PAN), are widely used in nanocomposite membrane production [18-21]. Single-walled carbon nanotubes compared to multi-walled carbon nanotubes have higher length

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to diameter ratio; therefore, they have higher application potential [22].

The most important issue regarding the application of nanocomposite membranes is the investigation of morphological features such as diameter, matrix structure, and permeability of the matrix bed [23]. In order to produce membranes containing CNTs and fibers with sub-micron diameter, the easiest and most effective method is to use electricity process [23, 24]. Electrospun fibers can be used with functionalized additives and fillers as nanocomposite fibers [25] In electrospinning method, it is possible to modify nanofiber structure by adding some of nanostructure materials to achieve desirable and new properties. Additives can influence properties and interactions in the solution as well as diameter and morphology of nanofibers. CNTs are widely used as fillers for electrospun fibers [21, 26-30] that were identified by Iijima [31]. So far, various methods have been used to treat greywater and these methods include physical [31,33] chemical 33] physiochemical [34,35] and biological methods [36-40]. Investigating the conducted studies in this context shows that many of physical, chemical, and biological processes cannot satisfy necessary needs to reuse greywater[10] Therefore, the treated greywater by these methods could not provide standard quality criteria for quick recycling. On the other hand, none of the mentioned methods could make recycling and treatment of gray water as practical for quick recycling in consumption process.

The purpose of the present study was to determine the efficiency of nanocomposite membranes made by polyacrylonitrile polymers containing SWCN in decreasing gray water pollution indicators as well as quick recycling of gray water in consumption process.

MATERIALS AND METHODS

Materials

Pure and white color powder of polyacrylonitrile polymer was prepared from Isfahan's Polyacrylamide Company with the molecular weight of $1.5*10^6$ g /mol. Di-methyl-formamide (DMF) with the purity percentage of 99.9% and density of 0.945 g/ml were prepared from Merk Company in Germany. SWCNT was prepared from US Research NanomaterialsInc with the approximate diameter of 9-15 nanometers. The analysis of SWCNT's main components by X-ray Diffraction Spectroscopy showed that these nanotubes contain 96.30% of pure carbon. Physical and morphological properties of these nanotubes are shown in Fig.1.

Preparation of CNT dispersions in PAN matrixes

To prepare PAN solution containing 1.82% and 2.5% CNT, first, PAN polymer was solved in DMF in controlled condition and temperature of 80 °C by magnetic stirrer for 30 minutes. Also, SWCNT were solved in DMF for 30 minutes at the temperature of 80 °C. In the next step, after mixing PAN and CNT to create uniform distribution of CNT in PAN, ultrasonic homogenizer machine (1500 W) with 50% power and timing of 3 seconds working and 7 seconds resting was used for 30 minutes in controlled condition.

Electrospinning process

According to previous studies [41-43], PAN polymer solution with 0-2.5% CNTs was prepared. In this study, Full Option Lab. ES electrospinning device made by Nanoazma Company was used. In this device, 5 ml syringe was used to inject polymer solution with robotic controlled infusion pump with the infusion rate of 1 cc per hour and 100 RPM with nuzzle diameter of 18 Gage. Other parameters of this process are shown in Table 1. Since the electrospinning chamber is surrounded, flow and air pressure measurements were ignored.

After producing synthesized nanocomposite membranes, the morphological structure of nanofibers produced by SEM images was evaluated after coating a thin layer of gold with an electron microscope (KYKY-EM3200). Figures 3A, 3B, and 3C show morphological structure of synthesized nanocomposite membranes.

Material	SWNT	MWNT	Steel
Young's modulus (GPa)	1054	1200	208
Tensile Strength (GPa)	150	150	0,4
Density (g/cm^3)		2,6	7,8
Thermal Conductivity W/m.K	3000		
Electrical Conductivity S/m	$10^5 – 10^7$		

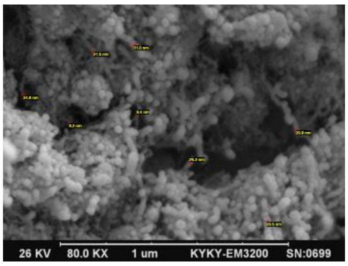

Fig.1. SEM image of SWCNT with the physical properties.

Table 1. Parameters in elecrospinning process of synthesized nanocomposite membranes

Parameters in elecrospinning process /type of synthesized nanocomposite membranes	Pure PAN	PAN+1.82% SWCNT	PAN+%2.5 SWCNT
Voltage (Kv)	20	11.6-15	22
Distance between nozzle tip to collector (cm)	9	8	21
Injection rates (ml per hour)	1.1	1	1.3
The horizontal movement collector (Scan) cm	3	2	5

Table 2. Maximum, minimum, mean, and standard deviation values of primary parameters as well as final parameters.

Parameter	Initial grey water samples(n=10)			Mixed Greywater	References no. Standard method[44]
	Min-Max	Mean	Standard Deviation		
Temperature ^{0C}	28-37	31.30	2.91	25	-
pH	6.9- 8.9	7.850	0.72	7.85	2520
Turbidity $_{NTU}$	38-109	79.60	27.02	73	2130A
COD $_{mg/L}$	110-645	312.80	167.10	258	5250C
BOD_5 $_{mg/L}$	52-294	141.60	75.92	135	5210A
TSS $_{mg/L}$	26-94	58.70	23.72	58	4500H
TDS $_{mg/L}$	49-301	180.00	91.53	155	1030
Detergent $_{mg/L}$	0.56-1.86	1.13	0.41	1.13	5540C

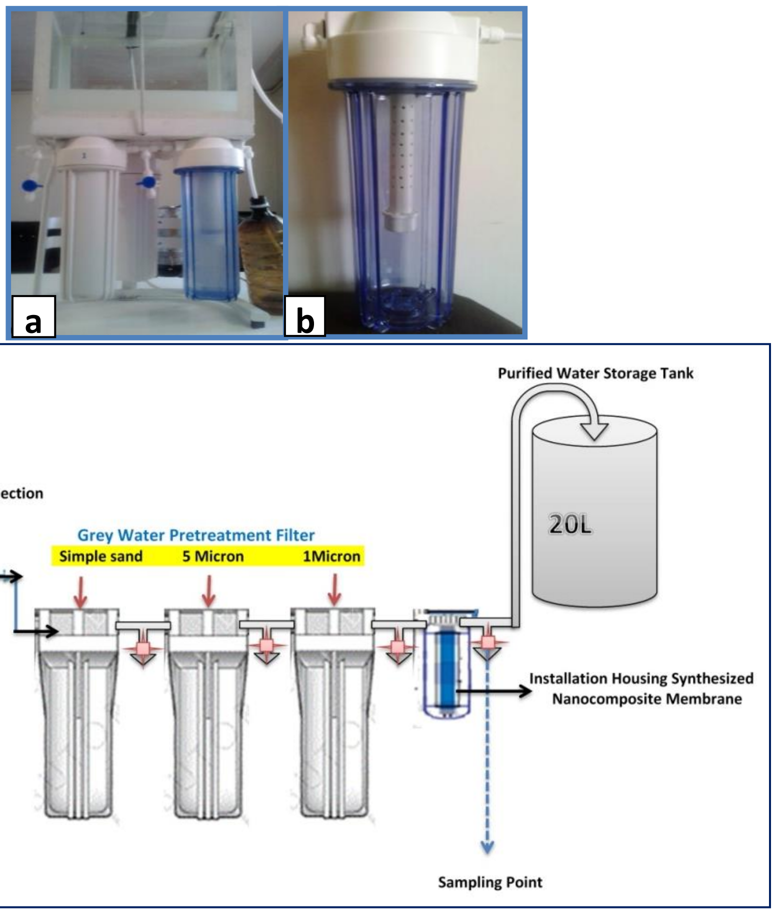

Fig. 2. a) Pilot device in this study b) Chamber for nanocomposite membranes installation c) Schematic of greywater treatment pilot equipment.

Sampling process

The greywater of 5 private bathrooms and 5 public bathrooms were collected (3L) and stored at the temperature of 4°C and transferred to the laboratory. In the laboratory, all samples were mixed in a big plate. All 10 primary samples were taken and the final sample was examined regarding chemical and biological parameters based on standards methods (44). Table (2) shows physiochemical properties of the primary samples and the final sample. To determine the efficiency of the membranes in the absence of biological factors, a synthetic solution was used. This solution was prepared from adding half McFarland ($1.5*10^8$), fecal coliform (E.Coli), and $2.8 * 10^2$total coliform to 1 liter physiological serum in OD=620nm.

Pilot design and launching

To examine the efficiency of synthesized membranes in the treatment and recycling of greywater, a pilot device was designed and launched. This device consists of a storage chamber of greywater, suction pump, and pilot infusion with 1.6 liters per minute flow rate ad optimized pressure of 130 PSI, Simple Sand Filters, 1 and 5 micron fiber filters and nanocomposite membranes installation that was used to install synthesized nanocomposite membranes with cross section of 0.05m2, 2(18*14 cm). Fig.2 shows the pilot device used in this study and Fiq.3 shows its schematic view.

Pilot testing

Twenty liters of the final sample size (30L) was picked to be stored in pilot chamber. During three steps, nanocomposite membrane installations were done. In each step, only one of the pure PAN, PAN+1.82% CNT, and PAN+2.5% CNT membranes were installed. In each step, 3L gray water was passed by 1.6L per minute under the pressure of 130 PSI from the pilot device (including filters and membranes) and the output wastewater was examined based on standard methods in Table (2) to determine the physiochemical parameters including COD, BOD, Turbidity, TSS, TDS, and Detergent to specify the efficiency of synthesized nanocomposite membranes in the treatment and recycling of greywater.

Bacteriology testing

To conduct testing in order to determine the efficiency of synthesized nanocomposite membranes in the absence of total and fecal coliforms, first, filters were sterilized in UV device and then, were installed at biological test pilot. Physiological serum with the concentration of half McFarland ($1.5*10^8$cfu/100ml) and total coliform ($2.8*10^2$cfu/100ml) added to the primary physiological serum, was passed through each membrane separately. Then, from the greywater passing through the membrane, final sampling was done in sterile glass (100cc). Samples were cultured based on 15-tubes MPN method (standard method 9221). First, in order to identify total coliforms in lactose medium, inoculation process was done at 35±0.5. After 48h, the initial results of positive and negative tubes were calculated based on table and Thomas formula. At the same time, with the culture of samples passing through membranes, control sample culture was done, too. In the final step, from 50% of positive carbonated tubes in the first step in the medium EC inoculation was done. After 48h in incubator at the temperature of 44.5 C, the presence of fecal coliforms was examined. The results of investigating the presence of total and fecal coliform bacteria available in control and final samples are presented in Table (3).

RESULTS AND DISCUSSION

The morphology of electrospun nanofibers (PAN+SWCNT)

In Fig.3, the structure of synthesized membrane nanofibers with statistical mean of nanofiber's diameter are shown. Mean and standard deviation of nanofiber's diameter produced by Pure Pan, PAN+1.82% CNT, and PAN+2.5% CNT were 867.75±318.25nm, 1131.7±445.39, and280.95±91.86nm, respectively. The results of this study are consistent with a study conducted in 2009 (41). To have more accurate investigation, the nanofiber diameter produced by SEM images with 500 x zoom was used (Fig.3). As can be seen from figures 3 and 4, the pore diameter in PAN+2.5%CNT is lower compared to Pure PAN and PAN+1.82%CNT.

Investigating the effect of key parameters on nanofiber electrospun

The effect of polymer solution concentration

With increased concentration of polyacrylonitrile polymer containing CNT, the mean of nanofiber diameter increases and this is due to increased physical entanglement of polymer chain 45)(. The physical entanglement of chains in the solution and increasing with concentration has led to increased viscosity of the solution and resistance against tensile force resulting from the available loads within the flow increases and finally leads to increased diameter. This is consistent with Deitzel et al. (45)

The effect of electric voltage: electric voltage is one of the effective parameters in electrospinning process of nanofibers. By increasing the voltage by 15 KW, the mean diameter of fibers increases and then decreases. On the other hand, with increased voltage, the electronic flow still increases. It is obvious that with increased voltage, due to increased homogenous loads, more flow passes through surface water in time unit and the surface density increases. Increased flow with increased voltage is consistent with Wan et al. .(46). In this study, voltage changes (11.6-22KW) were due to preventing bead formation at the tip of nuzzle and the surface of produced membrane surface as well as decreased wasted polymer solution (PAN+CNT).

The effect of distance between tip of the needle and collector: with increased distance between needle and collector, the electronic flow and the mean diameter decrease. Also, with increased distance, the electric intensity decreases and as a result, the jet speed decreases to towards the collector. Therefore, the flight duration increases and finally, leads to decreased diameter. On the other hand, if this distance is too much, due to the weakness of the electric field and lower strain, the mean diameter increases that can be observed at 20 cm .(47)

The effect of flow rate: with increased flow rate by 1 ml/h, the mean diameter of fibers increases and after that decreases. However, with increased flow rate, the electric flow does not follow a fixed trend. In fact, with increased infusion rate, the output volume increases while the voltage is fixed. But after that at 1.5ml/h flow rate, due to increased unspun drops under the influence of output solvent volume from the tip of the needle, the solution that is used for fiber production decreases and the mean diameter of the fibers decreases. The results of empirical observations that were mentioned above are consistent with Ramakrishna et al.(47) regarding the effect of flow rate on fibers' diameter. On the other hand, in higher flow rates and due to increased output solution volume from the tip of the needle, the drop corporations increase.

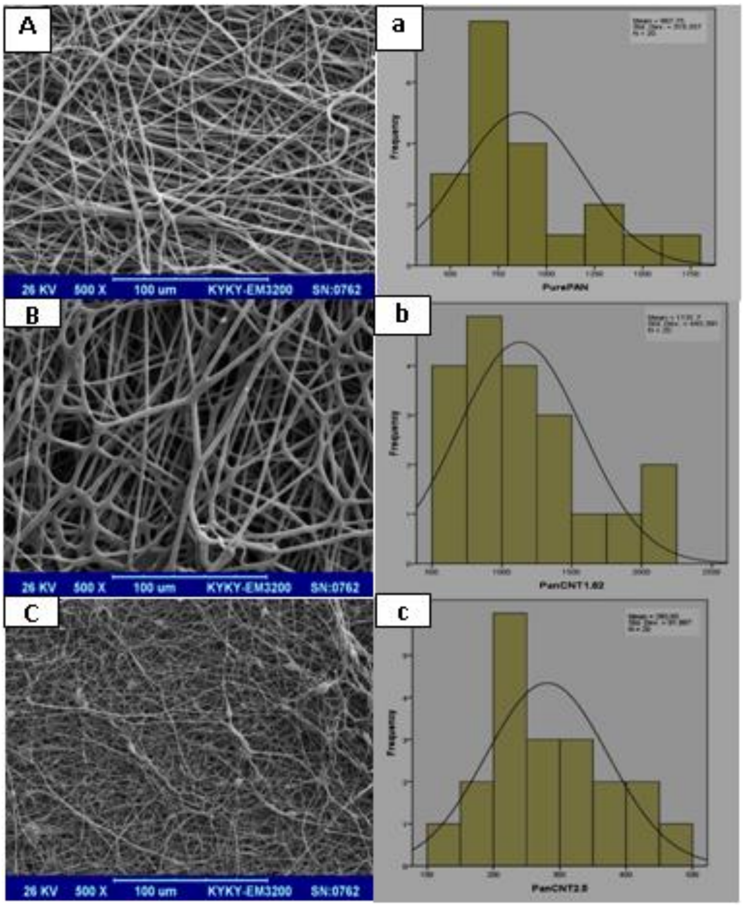

Fig. 3. A, B, and C show SEM images (500 x zoom) of Pure PAN polymer nanofibers, PAN+1.82% CNT, and PAN+2.5% CNT, respectively and a,b,c show nanofiber's diameter of Membranes a)- Pure PAN, b)- PAN+1.82% CNT, c)-PAN+2.5% CNT.

Table 3. Average influent and effluent grey water characteristics treated by pure PAN Nano composite membrane, PAN +1.82% SWCNT and PAN +2.5% SWCNT.(Q=1.6LPM, Hydraulic pressure=130 PSI .

parameter	Initial Influent grey water Avg.	Treated Effluent characterization /membrane type					
		Pure PAN		PAN +1.82% SWCNT		PAN +2.5% SWCNT	
		E. Avg.	R. P %	E. Avg.	R. P %	E. Avg.	R. P %
Temp.ºC	25	25	-	25	-	25	-
pH	7.85	7.2	-	7.6	-	7.5	-
COD, mg/L	258	69	73.25	117	54.65	25	98
BOD, mg/L	135	28	79.25	58	57	14	89.62
TSS, mg/L	58	17	70.68	24	58.62	5	91.4
TDS, mg/L	155	21	86.45	33	78.7	18	88.4
Detergent mg/L	1.1	0.5	54.5	0.6	54.45	0.1	90.9
Turbidity NTU	110	10	91	18	83.7	5	95.45
Fecal Coliform cfu/100ml	$1.5*10^8$	ND	100	ND	100	ND	100
Total Coliform cfu/100ml	$2.8 * 10^2$	<2	99.28	4	98.57	<2	99.28

E.Avg.: Effluent Average , R.P: Removal Percent %, ND: Not Detected

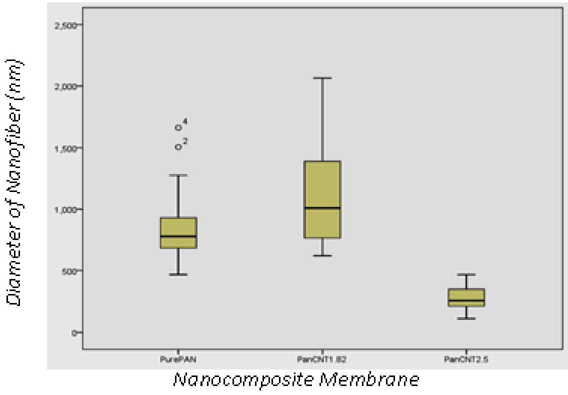

Fig. 4. Min, max and mean diameter of membrane nanofibers

In a study in 2015, it was shown that Pure Pan electrospinning leads to nanofibers production with the diameter mean of 515.34nm, but after adding 1% and 3% molecular weight of MWCNT, the structure and diameter mean of the produced nanofibers changed to 536 and 531nm, respectively [48]. It is consistent with the results of this study. Some scholars believe that the addition of CNTs to polymer matrix increases uniformity and mean of nanofiber diameter. The relative increase of the diameter can be due to increased viscosity of the final PAN/CNT solution [43]. However, in the present study, the addition of CNTs to the polymer matrix increased the nanofiber diameter and then, decreased it. This can be due to increased distance between nuzzle and the collector and also voltage and flow changes. The results of this study have relative correspondence with the results of the present study. In another study by adding Halloysite Nano Tubes (HNTs) to PAN in concentrations of 0, 1, and 3, improved electrospinning process and increased produced nanofiber diameter from 484nm to 556nm and 570nm were reported 49)(. Others reported the Pure PAN nanofibers diameters between 200 to 300nm and by adding 2%CNT to the primary solution, increased diameter to 500 and 600nm was reported 42)(that is consistent with the results of this study.

Analyzing biological and chemical experiments

In Table (3), the chemical and biological parameters, before and after the passage of greywater from synthesized nanocomposite membranes as well as the efficiency of these membranes in decreased indicators are shown.

All experiments were conducted at the standard temperature of 25 °C by applying error coefficient. The pH level after passing through three membranes based on Table (4), shows a little change.

The results of this study showed that PAN+1.8% SWCNT, Pure Pan, and PAN+2.5% SWCNT could decrease COD in primary greywater by 70.25%, 54.65%, and 98%, respectively. Also, BOD reduction percentages in water passing through these three membranes were 79.25, 57, and 89.62%, respectively. Therefore, the efficiency of PAN+2.5% SWCNT in COD and BOD reduction is significantly higher than other two membranes. The Pure Pan membranes compared to PAN+1.8% SWCNT showed more desired performance in decreasing these two indicators. By investigating the morphology of the mentioned membranes, it can be inferred that in the third membrane (Fig.3C), the pore diameter is very low among the nanofibers and this can be effective in attracting more pollutants. Also, due to increased pore diameter between the second membrane nanofibers (Fig.3B), the efficiency has been decreased.

In study by [36], RBC process was used to treat the greywater. In this study, the synthetic of biological reaction was investigated. The results of this study showed that the decline rates of COD and BOD were 84-89% and 64%, respectively [37].

Hocaoglu et al. [49] in a study using MBR process, in 60 days, reported BOD and COD removal efficiencies as 97% and 96.4%, respectively. The findings of this study are consistent with the results of the present study[50]. However, the mentioned results, due to increased treatment time and incapability of quick greywater recycling, are less important compared to the present study.

In another study, the use of ultrafiltration membrane led to decreased COD from 451mg/l to 117mg/l and BOD decreased from 274mg/l to 53mg/l [40]that compared to the present study and due to efficiency higher than 98% in the removal of COD and 89.6% in the removal of BOD, has better performance compared to ultrafiltration membrane.

The removal efficiency levels of greywater pollution indicators are shown in Fig.7 by each of the synthesized nanocomposite membranes.

In another study in 2013, the physiochemical processes were used to treat the greywater. The results showed the used process can decrease COD by 63%[51]. that compared to the present study has high weakness.

In another study where ultrafiltration, nanofiltration, and reverse osmosis membranes were used to treat the greywater, the efficiency of COD removal by ultrafiltration, nanofiltration, and reverse osmosis membranes were 53.5%, 93.36%, and 97.7%, respectively[52]. The results of this study are consistent with the results of the present study regarding the membrane and shows better performance compared to ultrafiltration membranes, but compared to reverse osmosis membrane, has lower efficiency that can be due to lower level of nanocomposite membrane in this study. In a study in 2004, the efficiency of nanofiltration membrane in removing the organic matters in greywater was 94% [53] that is a little bit better than the results of the present study. In another report in 2005, the reverse osmosis could decrease BOD from 82 to 2 mg that shows the efficiency of 98% that is more efficient compared to the present study regarding BOD removal [52]

In another study where ultrafiltration membrane with pore diameter of 0.05micron was used to treat greywater, BOD level declined from 195 to 86mg that shows 56% efficiency. The application of this membrane could not provide necessary standards for non-drinking uses of greywater [53,54] in this study [52] and compared to the results of the present study, shows lower efficiency in decreasing BOD. In a study in 1998 [40] by applying the relative ultrafiltration resistant membrane, BOD and COD levels declines from 451 and 271 mg/l in the input to 117 and 53 mg/l in the output that shows lower efficiency compared the results of the present study.

In a study in 2007, by implementing MBR with 0.01μm pores, the decline rates of COD and BOD were reported from 109 to 15 and 59 to 4mg/l, respectively [11, 46, 47].In a study in 2008, TSS-turbidity-BOD-COD declined from 29-43-23-55 mg/l to 9-4-9-22 mg/l 35)(that shows weaker efficiency compared to the present study. In another study in 2007, by implementing MBR membrane system, researchers could decrease COD by 96.7% and BOD by 95.7%[50]

TSS decline levels in output water from three mentioned membranes were 70.68, 58.62, and 91.4%, respectively. Also, the decline levels of TDS were 86.45, 78.7, and 88.4%, respectively. These results show that the efficiency of PAN+2.5% SWCNT is higher compared to other two membranes. This efficiency is significant in TSS decline. PAN membrane efficiency in TDS decline is at upper limit and is almost similar to PAN+2.5% SWCNT. The reason for high efficiency of PAN+2.5% SWCNT in removing TSS compared to TDS can be due to smaller pore diameters and the possibility for TDS penetration. Turbidity removal rate in PAN+2.5% SWCNT is high and determined as 95.45% that compared to other two membranes has higher efficiency. In several studies, the efficiencies of processes such as microfiltration and ultrafiltration in removing the organic matters (TDS and BOD) were not successfully evaluated, but about turbidity, TDS, and pathogens removals, acceptable performance was indicated. So that, turbidity and TSS removal efficiency were reported as 100% [40,52,53,56]

In 2013, in a study using MBR process and modelling the biological decomposition of greywater pollutants during 60 days, TSS declined from 51 to lower than 2mg/l (95.9%) [50] In a study in 2010, TSS and turbidity decline levels were evaluated as 83% and 90% using RBC process [37] and the findings of the present study show better efficiency compared to this study. However, in RBC method, time has a crucial role in greywater pollution decline; an issue that shows the superiority of the present study due to higher speed compared to other mentioned methods.

Also, in a study where ultrafiltration was used for greywater treatment, the final turbidity declined to NTU 1[57]. There is in a relative consistency with the present study.

In another study in 2007, by implementing MBR, researchers could decrease TSS from 51 to 2 mg/l (96%) [57].The results of this study are a little bit better that the present study regarding TSS removal and this can be due to increased utilization time from MBR.

In a study in 2013 where physiochemical processes were used to treat greywater, TSS removal was reported as 35% [51] that shows lower efficiency compared to the present study. In this study, the removal rate of linear surfactants (LAS) was 72% and in physiochemical processes, it was expected to have higher efficiency, but in comparison with the results of the present study, has lower efficiency**.** Detergents' removal rate in greywater by PAN+2.5% SWCNT nanocomposite membrane has the highest efficiency (90.9%), but in other two membranes, shows almost similar efficiency (54.5%) that can be due to large pore diameters compared to PAN+2.5% SWCNT.

Somewhere else in 2007, nanomembranes were used for greywater filtration that showed 92-98% of anionic surfactants and 88-92% non-anionic surfactants were removed by this membrane [58]. The results of this study regarding surfactants' removal are consistent with the present study.In another study, incapability of coagulation process in greywater treatment was reported. However, in this report, COD removal and anionic surfactants efficiencies were 70% and 90%, respectively [34]. The results of this study in microbial part showed that all three synthesized membranes have desired efficiency in removing pathogens ad fecal coliforms, so that all membranes can remove the fecal coliforms and this can be due to the large diameter of various *E. coli* bacteria between 0.2 to 2.4 micron (200-2400 nm [59]. It is larger compared to viruses with the diameter of 25 nm [10]. But in total coliform removal, PAN+2.5% SWCNT and Pure Pan showed similar efficiency (99.28%), but PAB+1.82% SWCNT showed lower efficiency (98.57%) in total coliforms removal.

In a study in 2010 using RBC process in greywater treatment, the coliforms removal by this process was 99.99% [37] that has a little bit higher efficiency compared to the present study.

In a study where ultrafiltration process was used for greywater treatment, *E. coli* removal rate was satisfying [57] .Is a study in 2008, total coliform and fecal coliform levels in all samples were lower than 1 in 100 ml[35]The results showed that fecal and total coliform decline are consistent with the present study. In another study where MBR process with the pore diameter of 0.03-1μm was used, the decline level of coliforms were 4-6 log [60]that is consistent with the present study. In 2007 ad by using MBR (0.01 μm) in the greywater treatment and recycling, it was shown that fecal coliform declined from CFU/100ml1.4×10^5 to 68 in 100 ml that is not consistent with global and state standards54)55 ,(. However, according to the country standards, it is suitable for agriculture [54] The results of this study([11]have lower efficiency in fecal coliforms decline rate compared to the present study. However, the required quality of the recycled greywater for each application according to the specific geographical zone is defined, but it should have the least criteria of organic matters, solids and biological requirements. One of the most important criteria is BOD5 lower than 10mg/l, turbidity lower than 2 NTU, and fecal coliforms at 100 ml [61]

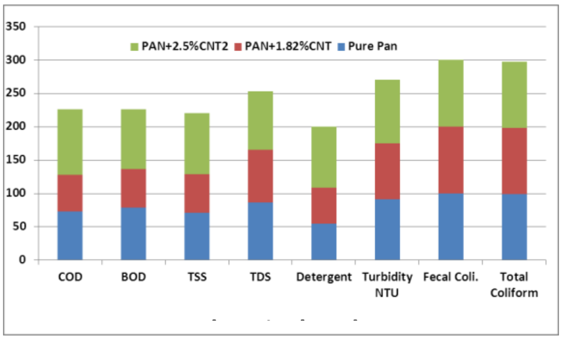

Fig. 7. Contribution of each treatment by Nanocomposit membrane synthesized to the removal of indicator pollutants from grey water

CONCLUSION

In sum, it can be concluded that according to the results obtained from this study, to dominate the most important challenges of water crisis in the future, modern technologies should be used to recycle water resources. One of the most important solutions can be the use of polyacrylonitrile polymer nanocomposites containing CNTs that have potential capability in removing greywater pollutants. Therefore, in near future, these fibers can be used to treat and reclamation of greywater in industrial scale. This can be effective step towards decreased water consumption per capita and water recycling.

REFERENCES

1. E. Eriksson, K Auffarth, M. Henze, A. Ledin *Urban water*, **4**, 85 2002.
2. B. Jefferson, J.E. Burgess, A. Pichon, J. Harkness, SJ. Judd, *Water research*, **35**, 2702 (2001).
3. R. Otterpohl, A. Albold, M. Oldenburg, *Water Science and Technology*, **39**, 153 (1999).
4. E.Eriksson, Potential and problems related to reuse of water in households. Ph. D. Thesis: Technical University of Denmark, Department of Environmental Science and Engineering, 2002.
5. P.A. Wilderer, *Water Science and Technology*.;**49**, 7 (2004).
6. L Allen, J. Christian-Smith, M. Palaniappan. Overview of greywater reuse: the potential of greywater systems to aid sustainable water management. Pacific Institute, 2010, p. 654.
7. D. Christova-Boal, R.E. Eden, S. McFarlane, *Desalination*, **106**, 391 (1996).
8. O.R. Al-Jayyousi, *Desalination*, **156**, 181 (2003).
9. F. Li, K. Wichmann, R. Otterpohl, *Science of the Total Environment*, **407**, 3439 (2009).
10. C. Merz, R. Scheumann, B. El Hamouri, M. Kraume, *Desalination*, **215**, 37 (200).
11. B. Jefferson, S. Judd, C. Diaper, Lens, P, Zeeman, G, Lettinga, G, Eds. , *Systems and Implementation*, **334**, 53 (2001).
12. D.A. Okun, *American Water Works Association Journal*, **89**, 52 (1997).
13. E. Santala J. Uotila, G. Zaitsev, R. Alasiurua R. Tikka, J. Tengvall, Microbiological greywater treatment and recycling in an apartment building. Proceedings of the 2nd International Advanced Wastewater Treatment, Milan, Italy. 1998, p.1416.
14. K.P. Lee, T.C. Arnot, D. Mattia, *Journal of Membrane Science*. **370,** 1 (2011).
15. M. Whitby, N. Quirke, *Nature Nanotechnology*, **2**, 87 (2007).
16. D. Mattia, K.P. Lee, F. Calabrò, *Current opinion in chemical engineering*, **7**, 32 (2014) .
17. D. Esrafilzadeh, M. Morshed, H. Tavanai , *Synthetic Metals*,.**159**, 267 (2009) .
18. M. Wu, Q. Wang, K. Li, Y. Wu, H Liu, *Polymer Degradation and Stability*, **97**,1511 (2012).
19. H.G. Chae, Polyacrylonitrile/carbon nanotube composite fibers: reinforcement efficiency and carbonization studies: ProQuest, 2008.
20. S. Prilutsky, E. Zussman, Y. Cohen, *Nanotechnology*, **19**, 165603 (2008).
21. W.J. Li, C.T. Laurencin, E.J. Caterson, R.S. Tuan, F.K. Ko, *Journal of biomedical materials research*, **60**, 613 (2002).
22. M.K. Pilehrood, P, Heikkilä, A. Harlin, *AUTEX Research Journal*, **12**, 1 (2012.
23. D.H. Reneker A.L. Yarin H. Fong , *Journal of Applied physics*, **87**, 4531 (2000).
24. H. Ye, H. Lam, N. Titchenal, Y. Gogotsi, F. Ko *Applied physics letters*, **85**, 1775 (2004).
25. Y.Q. Wan, J.H. He, J.Y. Yu, *Polymer International*, **56**, 1367 (2007).
26. S. Park, S. Jo, D. Kim, W. Lee, B. Kim, *Synthetic Metals*, **150**, 265 (2005).
27. H. Lam, N. Titchenal, N. Naguib, H. Ye, Y. Gogotsi, F. Ko, eds. Electrospinning of carbon nanotube reinforced nanocomposite fibrils and yarns. MRS Proceedings; Cambridge Univ Press ,.2003:
28. L. Vaisman, E. Wachtel, H.D. Wagner, G. Marom, *Polymer*, **48**, 6843 (2007).
29. Y-W. Ju, G-R. Choi, H-R,Jung W-J. Lee, *Electrochimica Acta*, **53**, 5796 (2008).
30. M. Ward, Treatment of Domestic Greywater Using Biological and Membrane Separation Techniques: MPhil Thesis, Cranfield University; 2000.
31. S. Prathapar, M. Ahmed, S. Al Adawi, S. Al Sidiari, *International journal of environmental studies*, **63**, 283 (2006).
32. E, Eriksson, K. Auffarth, A.M. Eilersen M. Henze, A. Ledin, *Waters*, **29**, 135 (2003).
33. Y. Chang, M. Wagner, P. Cornel, *Gewasserschutz Wasser Abwasser*, **206**, 32 (2007).
34. M. Pidou, L. Avery, T. J. Stephenson, P, Effrey S.A. Parsons, S. Liu, *Chemosphere*, **71**, 147(2008).
35. T.A. Elmitwalli, R. Otterpohl, Water Research. **41**, 1379 (2007).
36. A. Baban, S.M. Hocaoglu, E.A. Atasoy, K, Gunes, S. Ayaz, M. Regelsberger, *Desalination and Water Treatment*, **23,** 89 (2010).
37. M. Elmitwalli, C. Shalabi, R. Wendland Otterpohl, *Water Science and Technology*, **55**, 173 (2007).
38. L. Hernandez, G. Zeeman, H. Temmink A. Marques, C. Buisman eds.., in: Proc IWA conference on Sanitation challenges, Wageningen, Netherlands, 2008.
39. R.Birks, Biological aerated filters and membranes for greywater treatment: MSc thesis, Cranfield University; 1998.
40. P. Heikkilä, L. Wikström, A, Pasanen, P. Kauranen, A, Harlin eds., Preparation of electrospun pan/cnt composite fibres. (ICCM17 Edinburgh 17th international Conference on Composite Materials 17 Jul 2009-31 Jul 2009), Edinburgh, UK; 2009.
41. N. Khenoussi, E. Drean, L. Schacher, D. Adolphe, H.Balard, *Materials Technology*, **24**, 36 (2009).
42. G. Mathew, J. Hong, J. Rhee9, H. Lee, C. Nah *Polymer Testing*, **24**, 712 (2005).
43. A. Apha, WPCF, Standard methods for the examination of water and wastewater. American Public Health Association/American Water Works Association/Water Environment Federation, Washington DC, USA. 1995.
44. J.M. Deitzel, J. Kleinmeyer, D. Harris, N.B. Tan, *Polymer*, **42**, 172 (2001).
45. Y-Q. Wan, J, He, J. Yu, *Iranian Polymer Journal*, **15**, 265 (2006).
46. S. Ramakrishna, K. Fujihara, W-E Teo, T-C Lim, Z. Ma,World Scientific; 2005.
47. O. Eren, N. Ucar, A. Onen, N. Kizildag, Marmara J. Pure Appl. Sci., Special Issue-1: 95 (2015).
48. M. Makaremi, R.T. De Silva, P .Pasbakhsh, *The Journal of Physical Chemistry C*, **119**(14), 7949 (2015).

49. S.M. Hocaoglu, E. Atasoy, A. Baban, D. Orhon *Journal of Membrane Science*, **429**, 139 (2013).
50. C. Noutsopoulos, A. Andreadakis, N. Kouris, P. Mendrinou, I. Mantziaras, Physicochemical treatment of greywater, Proc. 14th Int. Conference on Environmental Science and Technology, Rhodes, Greece, 3-5 September 2015.
51. S. Šostar-Turk, I. Petrinić Simonič, M. *Conservation and Recycling*, **44**, 185 (2005).
52. G. Ramona, M. Green, R. Semiat, C. Dosoretz, *Desalination*, **50**, 170 (2004).
53. Organization WH. Guidelines for the Safe Use of Wastewater, Excreta and Greywater: Excreta and greywater use in agriculture: World Health Organization; 2006.
54. Epa U. Guidelines for water reuse. Environmental Protection Agency, Municipal Support Division Office of Wastewater Management Office of Water Washington, DC Agency for International Development Washington DC, EPA/625/R-04/108, Cincinnati, OH US EPA/625/R-04/108. 2012.
55. K-H. Ahn, J-H. Song, H-Y. Cha, *Water Science and Technology*, **38,** 373 (1998).
56. F Li, J. Behrendt, K. Wichmann, R. Otterpohl *Water Science and Technology,* **57,** 1901 (2008).
57. N. Funamizu, Y. Kikyo G*ewasserschutz Wasser Abwasser*, **206**, 35 (2007).
58. Nelson DE, Young KD. *J. Bacteriol*., **182**, 1714 (2000).
59. C. Chiemchaisri, Y. Wong, T. Urase, Yamamoto, *Water Sci. Technol.,* **25**, 40 (1992).
60. B. Jefferson, A. Palmer, P. Jeffrey, R. Stuetz, S. Judd, *Water Sci. Technol.,* **50**, 157 (2004).