Produced Water Treatment AIP

by Subriyer Nasir

AIP Conference Proceedings

Produced water treatment using integrated modified surface of kapok fibres and ceramic membrane

Cite as: AIP Conference Proceedings **2563**, 040010 (2022); https://doi.org/10.1063/5.0103182
Published Online: 31 October 2022

Subriyer Nasir, Sylvia Ramadhanti, M. Fawaz Rifki, et al.

Produced Water Treatment Using Integrated Modified Surface of Kapok Fibres and Ceramic Membrane

Subriyer Nasir[1,2, a)], Sylvia Ramadhanti[2], M. Fawaz Rifki[2], Herlina Wahab [3], Taufik Ari Gunawan [4]

[1]Laboratory of Separation and Purification Engineering, Faculty of Engineering, Universitas Sriwijaya. Jl Palembang-Prabumulih, Indralaya Ogan Ilir 30662 Indonesia
[2]Department of Chemical Engineering, Faculty of Engineering, Universitas Sriwijaya, Jl. Palembang-Prabumulih Ogan Ilir 30662 Indonesia
[3]Department of Electrical Engineering, Faculty of Engineering, Universitas Sriwijaya, Jl. Palembang-Prabumulih Ogan Ilir 30662 Indonesia.
[4]Department of Civil Engineering, Faculty of Engineering, Universitas Sriwijaya, Jl. Palembang-Prabumulih Ogan Ilir 30662 Indonesia.

[a)] Corresponding author: subriyer@unsri.ac.id

Abstract. The treatment of produced water from oil and gas fields before reinjection into reservoirs is an effort to reduce oil & grease include heavy metals that harmful to water bodies. In the current work, the modified kapok fiber (*Ceiba Pentandra. L*) surface using hot water for oil & grease removal was investigating on a laboratory scale. The present work aims to evaluate the modified kapok fibres surface (MKFS) using hot water as an adsorbent and commercial ceramic membrane in removal some contaminants of produced water, especially for barium ion. Some parameters of produced water determined were chemical oxygen demand (COD), total dissolved solids (TDS), oil & grease, and barium ion concentration. The effect of hot water treatment on the MKFS was evaluating using SEM-EDX. Results show that MKFS using hot water at 85 °C effectively removed 99.4% oil & grease, 12.2% TDS, 76.6% COD, and 38.7% barium from produced water. Result also shown that in the proposed method, the total removal of COD, TDS, oil & grease, and barium ions is 98.3%, 73.6%, 73.2%, and 76.4%, respectively. It concluded that the MKF surface is the potential method for diminished oil & grease and heavy metal from produced water.

INTRODUCTION

Produced water (PW) is liquid waste generated from oil and gas exploration, particularly for the enhanced oil recovery process. The PW contains hazardous chemicals that originate in oil and natural gas and from drilling activities. Pollution by PW caused by oil & grease, heavy metals, and total dissolved solids (TDS) are harmful to the environment [1-5]. Heavy metals that accumulate in water and soil, particularly in PW, are carcinogenic and very toxic [6]. At present, the PW is conventionally treated through different physical, chemical, and biological methods before reinjecting into reservoirs. Various treatments of PW are proposed by researchers, for instance, using a membrane [7,8], adsorption [9-11], electrocoagulation [12], and pervaporation distillation [13].

Currently, alternative low-cost natural adsorbents from agriculture and livestock are widely developing. For example, kapok fibres are used as the sorbent of oil & grease from produce water because of their characteristics such as hydrophobic, low-cost, biodegradable, and available in nature. Kapok is a cellulosic fiber with a high degree of hollowness (80–90 %), considered the largest among natural fibers [2,16]. Therefore, kapok fibres have been used as

The 2nd Universitas Lampung International Conference on Science, Technology, and Environment (ULICoSTE) 2021
AIP Conf. Proc. 2563, 040010-1–040010-11; https://doi.org/10.1063/5.0103182
Published by AIP Publishing. 978-0-7354-4237-5/$30.00

an absorbent for oil spread on the water surface and to absorb the heavy metal contaminants in liquid waste [14-16]. Surface modification of natural fibres is an alternative to increase their sorption ability. However, among the available methods, low-cost and environmentally friendly materials for surface modification are considered when the sorbents are applied on an industrial scale. The current work implemented a combination of PW treatment using modified kapok fibres surface (MKFS) using hot water and followed by a commercial ceramic membrane at the end of treatment. The MKFS using hot water is an alternative for removing oil & grease and heavy metals from PW. It found that barium ions in PW can partially be removed using kapok fibres. In addition, the use of commercial ceramic filters for PW treatment effectively increases the TDS removal from PW. The results of the PW treatment by the proposed method would give recommendations for oil and gas companies to consider the use of the MKFS for oil & grease and barium removal.

MATERIALS AND METHODS

Materials

Produced water was supply by one of the oils and gas companies in South Sumatra, Indonesia. Produced water was collected and placed in a vacuum truck with capacities of 6000 L and sent to the laboratory. Kapok fibres (20 kg) were purchase in a local market in Palembang, South Sumatra, Indonesia. The installed equipment of PW treatment was a sand filter (SF) column, the MKFS absorbent column, and a ceramic membrane. The fiber-reinforced polyethylene (FRP) cylindrical tube with an inner diameter of 25 cm and 1350 cm of height was applied for the SF and acted as an adsorbent column. The sand filter column filled with 45 kg of sand at the bottom and 10 kg of active carbon at the top of the column. A series of three polypropylene sediment filters (pore size was 0.1, 0.3, and 0.5 µm, respectively) was installed as a pretreatment before fed into the MKFS column and a ceramic membrane. In the experiment, a ceramic membrane used produced by Doulton. The dimension of the ceramic membrane was 5 cm in thickness, 5 cm in width, and 25 cm in length. The Doulton ceramic membrane claimed has an average pore diameter of 0.1 µm. The pH was measured using a Horiba Laqua type PC210K pH meter and the Eutech TN100 turbidity meter used for turbidity measurement. The atomic absorption spectrophotometer AAS-ICAP 7000 series employed for determining the heavy metals. Heavy metal on the surface of MKFS characterized using Scanning electron microscope (SEM-EDX) type Vega.

Methods

There are three stages of filtration carried out in the current work. The first stage is filtration using silica sand and activated carbon, the second stage is adsorption using the MKFS, and the last stage is filtration using a ceramic membrane. Kapok fibres samples and deionized water used to prepare the MKFS using hot water. Kapok fibres were dried in the sunlight for 24 h and soaked in the deionized water at 85 °C for 45 min. Then kapok fibres were rinsed using clean water for color removal caused by impurities and dried in sunlight for two days. At the last step, kapok fibres were dried in the oven at 110 °C for two hours. Produced water from the sample tank transferred using a centrifugal pump at a flow rate of 3, 5, and 7 L min^{-1} to the sand filter column. The sample and the outlet product of the sand filter column, MKFS column, and the ceramic membrane are taken at the sampling point using a 1000 mL graduated glass at every 15 min of contact time. The total dissolved solids (TDS), chemical oxygen demand (COD), oil & grease content, and heavy metal were analyzed using APHA dan SNI (Indonesian National Standard). The experiment was started by measuring the initial parameters of produced water and compare to the results after treatment. The COD, TDS, oil & grease, and barium removal is calculating using the following equation.

$$\text{Removal (\%)} = \frac{\left(C_{in} - C_{out}\right)}{C_{in} \times 100} \tag{1}$$

Where C_{in} denotes the inlet concentration of the sample and C_{out} represents the outlet concentration of filtrates. The schematic of the laboratory experimental is showed in Figure 1.

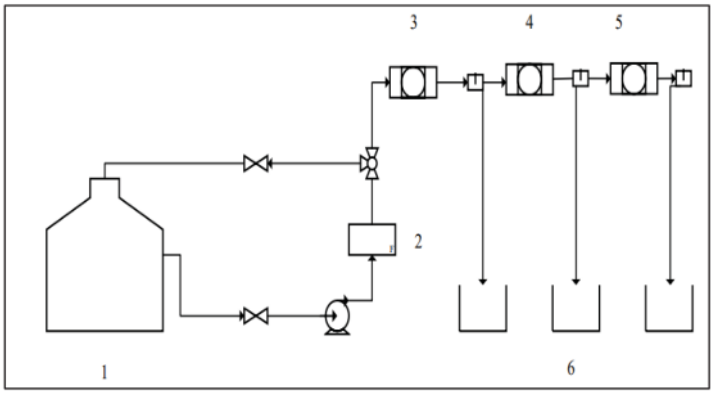


FIGURE 1. Scheme of produced water treatment

(1. Produced water tank, 2. Flowmeter 3. Silica sand filter 4. The modified kapok fibres surface sorbent 5. Ceramic membrane 6. Sampling tank)

RESULTS AND DISCUSSION

Produced water characteristics

The Physico-chemical characteristics of PW is illustrated in Table 1. The TDS, COD, oil & grease, and barium concentration of PW have exceeded the limit as stipulated by the Indonesian Ministry of Environment Regulatory No. 05/2014.

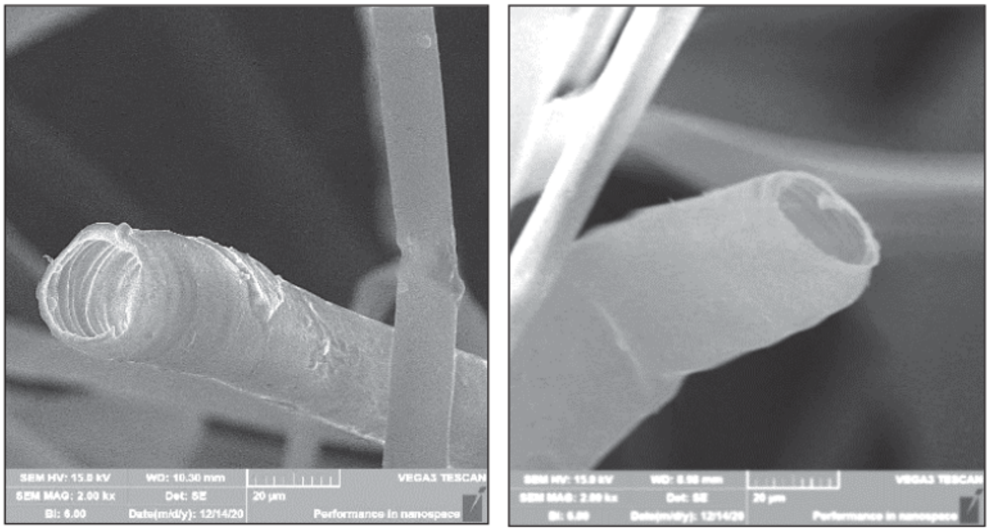
TABLE 1. Characteristics of produced water

No.	Parameter	Unit	Standard	Value
1.	pH	-	8-9	8.78
3.	TDS	mg L^{-1}	2000	14,700
4.	COD	mg L^{-1}	100	983
5.	Oil and grease	mg L^{-1}	10	18.3
6.	Barium	mg L^{-1}	2	5.84

Kapok fibres surface modification

Figure 2 shows the morphology of kapok fiber and MKFS using SEM at a magnification of 2000X. The raw kapok fibres are mainly composed of cellulose. They have a hollow structure called the lumen with a smooth wax layer on their surface. The presence of wax on the kapok fiber surface increased the hydrophobicity of cellulose. Hydrophobicity is related to the contact angle between kapok and water. The larger contact angle increases the hydrophobicity and the diffusion of heavy metal and oil & grease into the lumen. Previous research reported that kapok fiber from Java, Indonesia has a contact angle of 151.20. It is known for the super-hydrophobicity of kapok fiber.

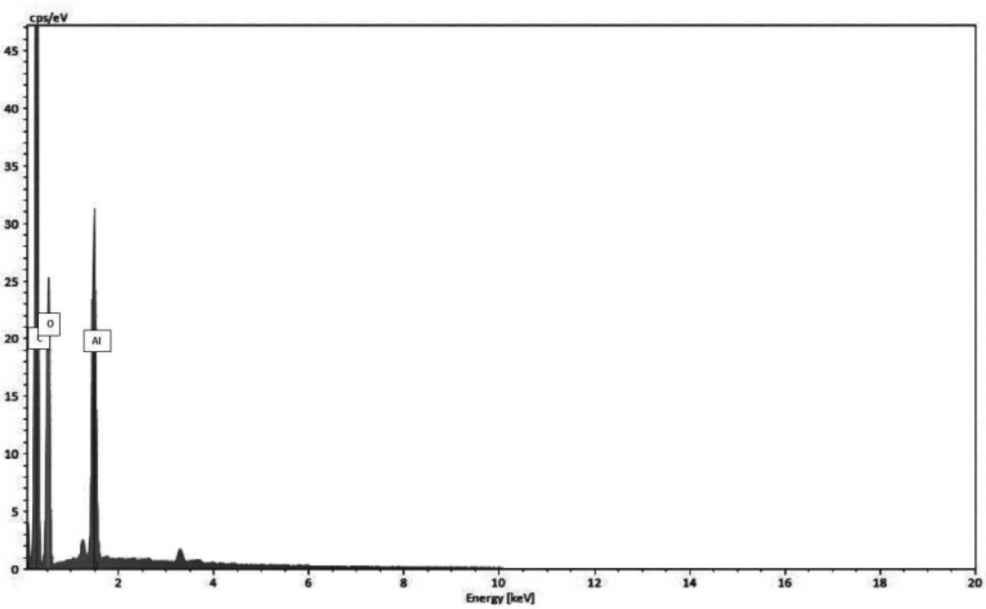
It suggests that oil & grease, and metal ions diffusion occur on the internal voids of the lumen through the Van der Waals force, capillary action, and hydrophobic interaction [17]. The wax layer roughness of MKFS plays a role in absorbing the metal ions and oil of produced water. Treatment of kapok fibres using hot water increasing the roughness of their surface and decreasing the hydrophobicity. Thus, the ability of kapok fiber in absorbing oil & grease and metal ions will increase. This result was similar to previous research using chloroform and ethanol for kapok fiber surface modification [18].



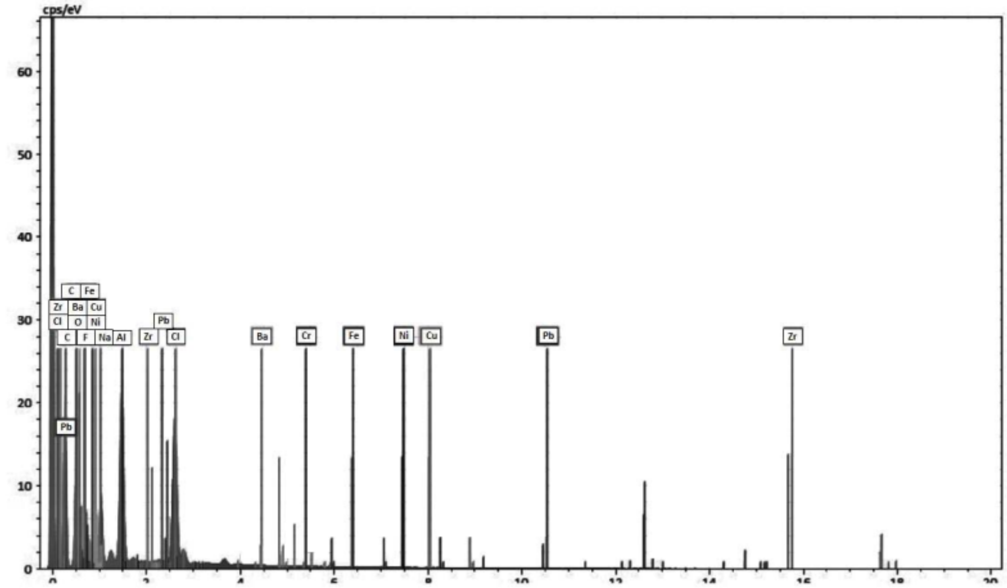
(a) (b)

FIGURE 2. SEM images of a) raw kapok fibres and (b) the MKFS

The modified surface of kapok fibres has shown the ability to absorb the metal ions including Na, Al, Ni, Fe, Ba, Cu, Pb, Cr, and Zr as illustrated in Figure 3. The EDX analysis showed that the surface of modified kapok fibres with hot water increased the lumen capillary activity and its hydrophobic properties and affected the oil absorption capacity [19].



(a)



(b)

FIGURE 3. The EDXs of a) raw kapok fibres and b) treated kapok fibres

Figure 3. show the EDX spectra of kapok fibres Kapok fibres is cellulose Elements such as C, O, and Al are components of kapok fibres. Table 2 illustrates the presence of metal elements such as Fe, Ni, Cu, Cr, Zr, and Ba on MKFS indicates that kapok fibres potentially be applied as metal ion absorber of the PW. It is also showed the presence of barium with the concentration exceeding the regulation. The MKFS has shown the capability to absorb the metal ions as illustrated in Figure 3. The EDX analysis showed that the MKFS using hot water increasing the lumen capillary activity and its hydrophobic properties and affected the oil absorption capacity [19]. Previous research on leaf moss modified surface with hot water at 80 °C for absorbing oil is similar when using alkali as the solvent [20]. From an economic point of view, hot water will be attractive for surface modification of kapok fibres compare to use of chemical solvents.

TABLE 2. Elemental composition of modified kapok fibres surface

Elements	Composition (mass %)	
	Before absorption	After absorption
Carbon	55.01	50.44
Oxygen	39.06	34.87
Aluminum	5.93	5.20
Chlorine	-	4.99
Sodium	-	3.91
Fluorine	-	0.47
Nickel	-	0.03
Iron	-	0.02
Barium	-	0.02
Copper	-	0.02
Lead	-	0.01
Chromium	-	0.01
Zirconium	-	0.01

Total Dissolve Solids

The TDS value is one of the water quality parameters representing the total concentration of the dissolved substance in water. This term refers to inorganic salts and a small amount of organic material dissolved in water naturally or due to industrial activity. Produced water sample use in this research has a high TDS (ca 15,000 mg L^{-1}). A high TDS will affect the organoleptic properties of water and also disturb the environment.

Figure 4 shows TDS removal of PW in the pretreatment unit at the range of contact time 0-120 min and flow rate of sample from 3 L min^{-1} to 7 L min^{-1}. The optimum TDS removal is 8.33% at the sample flow rate of 5 L min^{-1} and contact time 120 min. The average TDS removal in the pretreatment unit is 14.59%. The results showed that TDS removal decreased with increasing contact time and flow rate of the sample. It is suggested that the pretreatment system is clogging due to solid particles precipitation. Silica sand and sediment filter in the pretreatment aims to remove various kinds of dissolved solids in produced water. Activated carbon can remove organic and inorganic substances, while silica sand can absorb suspended solids in produced water [4]. The silica sand, activated carbon, and sediment filters have a significant role in TDS removal. Therefore, the use of these components is suitable for removing the solid particle from water and wastewater. Activated carbon is helpful in the adsorption of not only color but also dissolved solids in water.

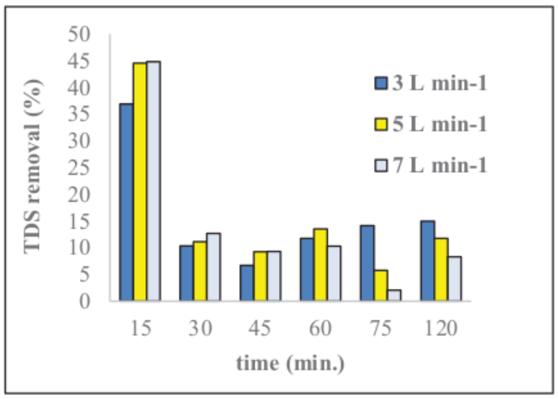


FIGURE 4. TDS removal of the pretreatment filtrates at different flow rate

The different results are found in the use of MKFS to reduce dissolved solids shown in Figure 5. The optimum removal of TDS using MKFS is 12.77% and a minimum of 3.38% at flow rate of 5 L min^{-1}. The average TDS removal of MKFS is 8.89%. This value is lower than those of the pretreatment result. The previous study has also shown that kapok fibres can reduce the concentration of metal ions and oil because of the interaction of kapok fibres wax and oil surface is controlled by the van der Waals forces [17, 21]. The mobility of dissolved solid particles is affected by flow rates of PW sample. An increase of flow rate and contact time will increase the mobility of solid particles and lead to optimal dissolved solid absorption. However, the removal of contaminants in wastewater treatment not only affected by flow rate of feed but also gravitational force [22].

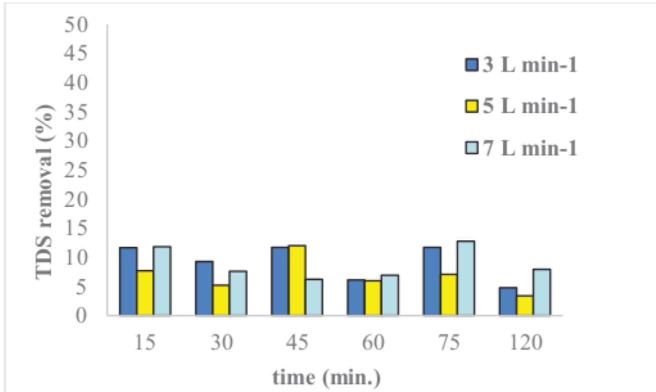


FIGURE 5. TDS removal of MKFS filtrates at different flow rate

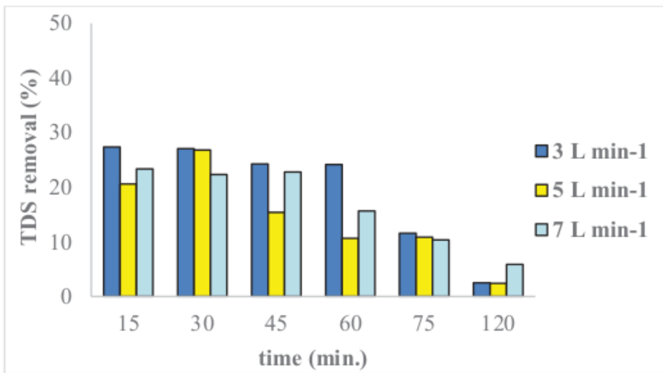


FIGURE 6. TDS removal of ceramic membrane permeates at different flow rate

Figure 6 shows the TDS removal of ceramic membrane permeates at a different flow rate. The TDS removal of permeate is 26.82% at 30 min contact time and continues to decrease to 2.41% after two hours of operation at a flow rate of 5 L min^{-1}. with the average TDS removal of ceramic membrane is 16.73%. Increases in the contact time and flow rate are affected the TDS of permeates. The TDS consists of the amount of inorganic and organic substances in water. This substance can also be in the form of suspended particles. The dissolved solids and suspended solids retained on the ceramic membrane surface may cause the membrane fouling by inorganic matter because ceramic membranes are less prone to organic fouling due to their hydrophilicity. The inorganic substance in the PW sample suggested increasing the TDS values. Therefore, the application of pretreatment and the uses of MKFS combined with ceramic membrane improve the removal of organic and inorganic pollutants.

In the current work, the TSS effect on turbidity removal is related to turbidity removal. An increase in TSS will decrease turbidity removal. It suggested that the permeate is relatively free of TSS due to the pretreatment process and the use of the MKFS before feeding to the ceramic membrane. Generally, the particle size of the suspended solids in water is 2 µm and is much larger than the average pore diameter of the ceramic membrane, which is 0.1 µm. Therefore, the dissolved solids will be concentrated on the membrane surface. The ceramic membrane used in the experiment is 0.1 µm and may categorize as a microfiltration membrane. Therefore, practically not only TDS but also total suspended solid (TSS) will be retained on the membrane surface. Although the total suspended solid is not measure in the experiment, their effect on the turbidity of water is significant. Figure 7 shows that permeate turbidity removal of the ceramic membrane reaches 94%.

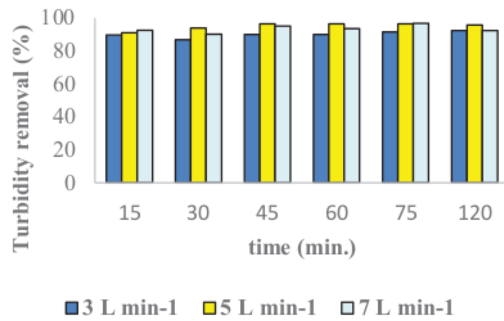


FIGURE 7. Turbidity removal of ceramic membrane permeates

Chemical Oxygen Demand (COD)

In this study, the PW sample at a flow rate of 5 L min^{-1} and contact time of 75 min was selected for COD, oil & grease, and barium concentration analysis because it has an optimum pH and turbidity. The results obtained compared to standards oil and gas industrial wastewater. Figure 8 show the COD removal of the pretreatment unit, the MKFS, and the ceramic membrane is 68.87%, 76.60%, and 88.30%, respectively. The pretreatment process and the MKFS have a high COD removal, but the results still exceed the standards. The COD of the ceramic membrane permeate decreased from 983 mg L^{-1} to 115 mg L^{-1} and has met the Indonesian standard.

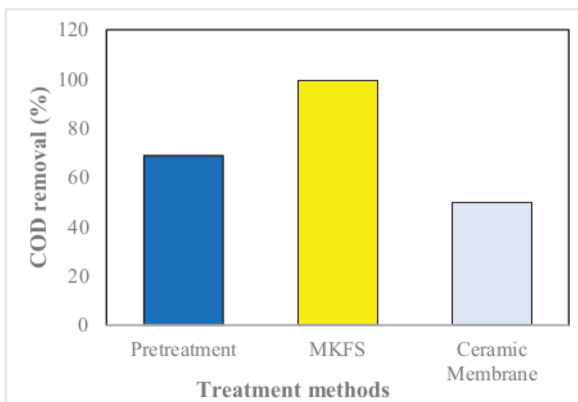


FIGURE 8. COD removal at flowrate of sample 5 L min^{-1} and contact time 75 min.

A large amount of organic and inorganic components in water can cause COD values to increase [4]. The use of additional chemicals during the drilling process may reduce dissolved oxygen levels and increase organic and inorganic substances of PW. The average COD removal for the current propose method is 88.30%.

Oil and grease

Figure 9 illustrates the oil & grease removal of the pretreatment, the MKFS, and ceramic membrane after two hours of contact time. The oil & grease removal for the pretreatment system is only 69.95%. This is because of the presence of sand which filtered the oil & grease from PW. However, more than 99% oil & grease removal is obtained in MKFS and 50% in ceramic membrane. The average oil & grease removal in the current work is 73.2%. The use of MKFS in PW treatment was able to increase oil absorption up to 30% because the acetyl group in the kapok fibres is hydrophobic even after wax removal. The hydrophobic-oleophilic characteristics of the kapok fiber could be attributed to its waxy surface, while its large lumen contributed to its excellent oil absorbency and retention capacity.

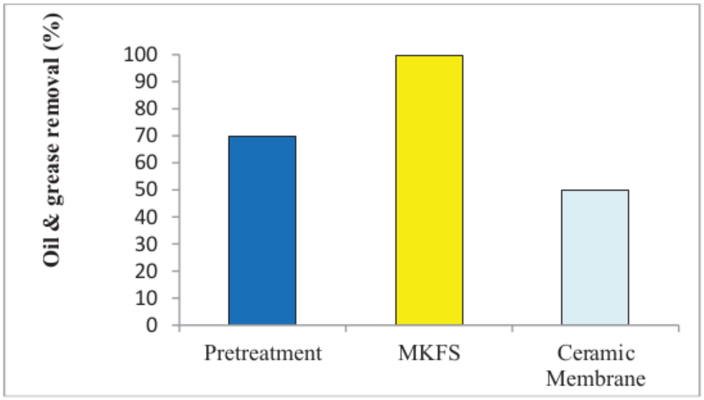


FIGURE 9. Oil and grease removal

Barium concentration

The world oil and gas companies produced the PW with barium concentration from 1.3 to 650 mg L^{-1} depending on the geographical features, age and chemical composition of the reservoirs, and methods of extraction [4]. The high concentration of barium ion in PW could give a strong indication of the presence of radium isotopes [23]. Therefore, oil and gas companies should pay attention to a high concentration of barium of PW which is harmful to the environment. One method applied by companies is re-injected the produced water into oil wells. Problems will arise when barium is carried away by the water flows into water bodies. The radioactivity effect on humans cause by radium must also be taken seriously.

Figure 10 illustrated barium removal for different treatment methods. The total concentration of barium in PW is 5.84 mg L^{-1}. In the pretreatment unit, barium removal is about 8.3%. It indicates that a small amount of barium metal only adsorbs by active carbon in the sand filter column. The barium removal for MFKS and the ceramic membrane is about 32.9 and 38.44%, respectively. The EDX spectra of MFKS has confirmed the presence of barium ion. In addition, not only barium but also heavy metals such as iron, copper, nickel, lead, and chromium retained on the surface of kapok fibres. Increasing of flow rate will increase the concentration of barium on the lumens. The mechanism of deposition of heavy metal on the surface of kapok fiber is due to the lumen capillary activity and diffusion [17,19]. Barium and other heavy metal ions deposition on ceramic membrane pores may occur via solution diffusion mechanism and support by the hydrophilicity of the ceramic membrane. Overall, the average total barium removal of PW in the current experiment is 76.4% and meets the requirements of PW reinjection into reservoirs or discharge to water bodies.

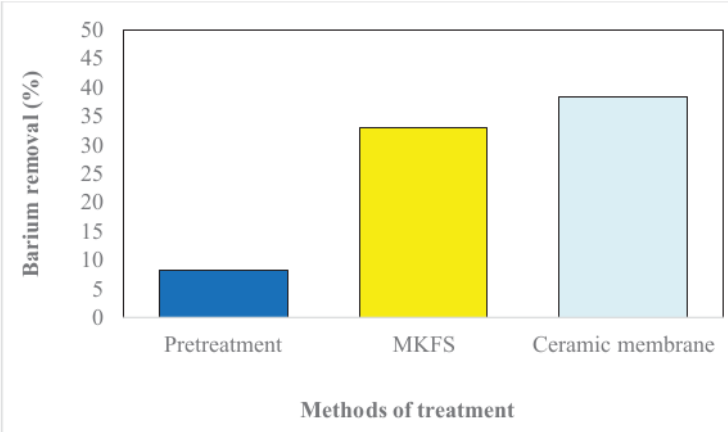


FIGURE 10. Effect of treatment methods on barium removal

Table 3 compiled the result of final treatment of PW. The results show that the propose method only decreased the COD, oil & grease, and barium concentration. It can be seen that the final pH was 8.15. It means there is a lower decreased on acidity (7.18%) and the permeate is still alkaline. However, the TDS is still high and do not meets the regulation.

Table 3. **Produced water characteristics at the end of treatment**

Parameter	Unit	Pretreatment	MFKS	Ceramic Membrane
pH	-	8.27	8.21	8.15
COD	mg L^{-1}	306	230	115
TDS	mg L^{-1}	8150	7150	5230
Oil & grease	mg L^{-1}	5.5	0.02	<0.010
Barium	mg L^{-1}	5.35	3.59	2.21

CONCLUSION

The modified kapok fiber's surface using hot water increases the surface roughness of kapok fibres. As a result, increase the oil & grease removal up to 99.9% and remove barium ion of produced water by 38.7%. The modified kapok fiber's surface using hot water is effectively absorbs heavy metals such as nickel, iron, copper, lead, chromium, and zirconium from produced water. Treatment of produced water using the combination of pretreatment using silica sand/activated carbon, modified kapok fibres surfaces, and ceramic membranes showed the total removal for COD, TDS, oil & grease, and barium ion is 98.3%. 73.6%, 73.2%, and 76.4%, respectively. The proposed method can be applied in oil and gas reservoirs for produced water re-injection into the reservoirs.

REFERENCES

1. A. Fakhru'l-Razi, A. Pendashteh, L.C. Abdullah, D.R.A.Biak, S. S. Madaeni, and Z. Z. Abidin, J. Hazard. Matter. **170**, 530-551 (2009).
2. S. Jiménez, M. Micó, M. Arnaldos, F. Medina, and S. Contreras, Chemosphere **192**,186-208 (2018).3
3. M. Al-Kaabi, M. Al-Ghouti, M. Ashfaq, T. Ahmed T, and N. Zouari, J. Water. Proc. Eng. **31**, 100830 (2019).
4. M. Al-Ghouti, M. Al-Kaabi, M. Ashfaq, and D. Da'na, J. Water Proc. Eng. **28**, 222-239 (2019).
5. J. Wang, and C. Chen, Biotechnol. Adv. **27**, 195–226 (2009).
6. S. Munirasu, M. Haija, and F. Banat, Process Saf. Environ. Protection **100**,183-202 (2016).
7. J. Dickhout, J. Moreno, P. Biesheuvel, L. Boels, R. Lammertink, and W. M. de Vos, J. Colloid. Interf, Sci. **487**, 523-534 (2017).
8. B. G. P. Bezerra, A. Parodia, D. R. Da Silva, and S. B. C. Pergher, J. Environ. Chem. Eng. **7**,103006 (2019).

9. Z. Ye and V. Prigiobbe, Water. Research **185**,116258 (2020).
10. L.T. Hendges, T.C. Costa, B. Temochko, S.Y.G. González, L.P. Mazur, B.A.Marinho, A.Silva, S.E. Weschenfelder, A.A.U. de Souza, and S.M.A.G.U. de Souza, Process Saf. Environ. Prot. **145**, 262-272 (2021).
11. E. H. Ezechi, M. H. Isa, S. R. M. Kutty, and A. Yaqub, Process Saf. Environ.Prot. **92**, 509-514 (2014).
12. J. Wang, D. Tanuwidjaja, S. Bhattacharjee, A. Edalat, D. Jassby, and E. M. V. Hoek, Water **12,** 3560 (2020).
13. T. Dong, G. Xu and F. Wang, J. Hazard. Mat. **296**,101-111 (2015).
14. X. Zhang, C. Wang, W. Chai, X. Liu, Y. Xu, and S. Zhou, J. Chem. Technol. Biotech. **92**, 1613-1619 (2017).
15. M. J. P. Macedo, G. S. Silva, M. C. Feitor, T. H. C. Costa, E. N. Ito, and J. D. D. Melo, J. Mater. Res. Tech. **9**, 2467-2476 (2020).
16. C. S. Quek, N. Ngadi, and M. A. Ahmad Zaini, J. Taibah Univ. Science **14**, 507-512 (2020).
17. A. U. Naharudin, S. H. N. Shaarani, L. M. Rou, N. H. Hamidi, N. Ahmad, and R. Rasid, J. Chem. Technol. Biotech. **5**, 48-54 (2020).
18. E. Anuzyte and V. Vaisis, Energy Procedia **147**, 295-300 (2018).
19. S. Viju and G. Thilagavathi, J. Nat. Fibres **5,** 1-9 (2020).
20. T. T. Lim and X. Huang, Chemosphere **66**, 955-963 (2007).
21. N. Lv, X. Wang, S. Peng, L. Luo, and R. Zhou, RSC Adv. **8**, 30257-30264 (2018).
22. E.T. Igunnu, and G.Z. Chen, Int. J. Low- Carbon. Technol. **9**, 157-177 (2014).
23. J. Usman, M.H. D. Othman, A.F. Ismail, M.A.Rahman, J. Jaafar, Y.O. Raji, A. O.Gbadamosi, T.H. El Badawy, and K.A.M. Said, J. Mater. Res. Technol. **12**, 643-667 (2021).