Total Dissolved Solids, Phenol, and Barium Removals from Oilfield Produced Water Using Kapok Fibers and Ultrafiltration Membrane

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Total Dissolved Solids, Phenol, and Barium Removals from Oilfield Produced Water Using Kapok Fibers and Ultrafiltration Membrane

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Abstract

Oilfield-produced water treatment using raw Kapok fiber (RKF) and a modified surface of Kapok fiber followed by an ultrafiltration membrane was conducted to reduce total dissolved solids (TDS), phenol, and barium. Variables considered in the experiment were contact times (30, 60, 90 min.), the flow rate of samples (5, 6, and 7 L/min), and trans-membrane pressure of ultrafiltration membrane (0.25, 0.35, and 0.50 bar). Raw Kapok fiber and modified surface Kapok fiber were used to investigate the effect of Kapok fiber on total dissolved solids, phenol, and barium removals from produced water. The results showed that raw KF decreased the TDS, phenol, and barium by 51.81%, 62.63%, and 54.20%, respectively. Treatment of raw Kapok fiber column filtrate using ultrafiltration membranes (UF) showed the removal of TDS, barium, and phenol achieved 73.15%, 42.44%, and 79.45%, respectively, at a flow rate of 5 L/min, TMP of 0.25 bar, and contact time of 90 min. Modifying the Kapok fiber surface using sodium hydroxide solution (5 wt%) and hot water (98.5 °C) reduced the TDS, phenol, and barium to 57.32%, 65.83%, and 79.08%, respectively. Further, at the same operating condition, the modified surface of Kapok fiber followed by UF decreased 94.31% TDS, 84.20% phenol, and 56.23% barium, respectively. The results show that modification of the Kapok fiber surface followed by UF can be used to remove the TDS, phenol, and barium from produced water.

Keywords

Kapok fiber, natural absorbent, oilfield, produced water, ultrafiltration

1 Introduction

Produced water (PW) from oil and gas exploration contains various compounds such as oil and grease, hydrocarbon, and dissolved salts. High amounts of salts dissolved in PW are both cations and anions such as sodium, potassium, calcium, magnesium and barium; anions such as chlorine, sulphate, carbonate, bicarbonate, and silica [1, 2]. As a results, untreated PW is harmful and dangerous for humans and the environment [3–6]. To date, the production of PW has been estimated around 250 million barrels per day [6] and more than 99% was dispose-off into the environment [7]. Therefore, before being reinjected into an oil reservoir or discharged into the environment, PW should be treated using physical, biological, or chemical methods [8]. Three parameters of PW in South Sumatra Indonesia considered the primary pollutant are total dissolved solids (TDS), phenol, and barium because their concentration exceeds the standard.

The higher TDS in PW will cause scaling in the piping system and water reinjection problems in the reservoir.

Phenol is a semi-volatile compound with a concentration between 0.001 and 10,000 mg/L found in PW and also a significant pollutant in wastewater streams from the petrochemical (2.8–1,220 mg/L), coal mining (9–6,800 mg/L) and petroleum oil refineries (6–500 mg/L) [8-10]. Phenol is very dangerous and harmful and causes the excretion of dark urine, impaired vision, diarrhea, and a sour mouth.

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Toxic phenol levels for fishes are between 9 and 25 mg/L and, for humans, are in the range of 10–24 mg/L [11].

Barium is a metal element in PW that can be found in the form of barium sulfate, carbonate, and barium chloride. The presence of barium in PW will cause environmental and operational problems in the oil and gas industry. Barium can harm a water-desalination system through interactions with naturally occurring carbonate and sulfate anions to form mineral scale fouling [12]. It is also suggested that a high concentration of barium ions in the PW strongly indicates radium presence [13].

Kapok (Ceiba Pentandra (L.) Gaertn) is commonly found in Sumatra and Java, Indonesia. Kapok was chemically composed of 64% cellulose, 13% lignin, and 23% pentosan [14]. Kapok is considered a natural fiber with a high degree of hollowness (80-90%) which composed of two primary layers. The outer layer of raw Kapok fiber (RKF) is composed of cellulose microfibrils, which are orientated transversally to the fiber axis, while the inner layer is parallel to the fiber axis. The specific surface area of RKF is evaluated as 2.99 m^2/g. The volume of the pores with diameters of 2–40 nm accounted for about 80% of the total pore volume, and those with diameters over 40 nm accounted for about 20% [14–16]. Cellulose in the RKF is coated with lignin, which strengthens cellulose's structure. Besides, the presence of lignin in RKF can interfere with cellulose to bind with metal ions. For this reason, RKF must be delignified to remove the lignin content.

The RKF has attracted the attention of researchers to absorb oil from water and wastewater [17]. RKF and its composites can be used as an adsorption material for heavy metal ions and dyes in an aqueous solution with the right pre-treatment and surface modifications [18]. Previous research shows that treated kapok using sodium hydroxide and hot water can be used as an oil sorbent [19, 20]. In addition, modified cellulose-based adsorbent was selectively removed 75% of barium in the simulated wastewater [21].

Combined adsorption and membrane technology in water and wastewater treatment has increased since the last decade because of their better separation performance than using a single conventional separation method.

The current work attempts to increase the hydrophilicity of RKF by surface modification using sodium hydroxide and hot water. The modified surface of Kapok fiber (MSKF) is used to treat PW, in particular for removing TDS, phenol, and barium.

2 Materials and methods

2.1 Materials

The PW samples were provided by one of oil and gas companies located in South Sumatra, Indonesia. RKF purchased from a local market in Palembang, Indonesia. The RKF and MSKF adsorption column was a fiber-reinforced polyethylene (FRP) cylindrical tube with a height of 1350 cm and a diameter of 25 cm. The adsorbent column was filled with 5.5 kg of RKF, which was modified using sodium hydroxide (NaOH) solution (5 wt%) and then soaked in hot water at 98.5 °C and dried in the oven at 105 °C for 2 h. The commercial ultrafiltration hollow fiber membrane type UF 4040 (MWCO 50,000 Da, 4 m^2 of membrane area) consists of a polyvinyl chloride (PVC) hollow fiber membrane placed in a stainless-steel housing with dimension: 100 cm of height, 10 cm of diameter and 0.2 cm of thickness. The membrane can be operated at pH range 2 to 11, maximum TMP of 0.2 MPa and temperature 5–45 °C.

2.2 Methods

The current research focused on improving the quality of PW using adsorption method by KF and filtration using a membrane. PW treatment was conducted using two stages. The first stage is a pretreatment using RKF and MSKF sorbents to remove solid particles, phenolic compounds, and barium ions from the PW sample. The second stage is the treatment of RKF and MSKF filtrates using a UF membrane.

The samples of PW from the feed tank were analyzed for TDS, barium, and phenol to determine the initial concentration. The sample is pumped into the absorbent column at a different flow rate (5, 6, and 7 L/min) and contact time (30–90 min.). The filtrate from the absorbent column was collected in the UF feed tank, where the filtrate was transferred into the UF module. The transmembrane pressure (TMP) for the UF membrane was set as 0.25, 0.35, and 0.50 bar. Permeates were collected in a polyethylene tank and sent to laboratory for TDS, phenol, and barium analysis.

2.3 Analysis

Total dissolved solids were measured using Horiba Laqua PC220-K type TDS meter. Phenol was determined using Shimadzu UV-vis spectrophotometer UV-1900i. The analysis were conducted by adding 4-amino antipyrine dye into sample and measured the absorbance UV at wavelength 470 nm. Barium concentrations were measured using the APHA 3120:2017 method using Thermo Scientific iCAP 7000 Series ICP-Optical Emission Spectrometer. The scanning electron microscope (SEM) with Electron Diffraction X-ray (EDX) Tescan type Vega 3 were used to investigate the structure of RKF and their elements. Schematic of experiment details can be found in Fig. 1.

3 Results and discussion

3.1 Produced water characterization

Table 1 shows the PW analysis of acidity, total dissolved solids, phenol, and barium. TDS refers to organic and inorganic substances dissolved in water or wastewater; their concentrations in the PW vary from 100 mg/L to 400,000 mg/L [6]. The TDS, phenol, and barium concentration of PW samples do not meet quality standards for wastewater. Even when compared to produced water from other areas, the value of this parameter is still relatively low. However, produced water must be treated before being discharged into water bodies or reinjection into the oil wells.

3.2 Acidity

Table 2 illustrated the acidity of PW filtrate after adsorption by RKF and MSKF. There is no significant change of pH at varied flowrate and contact time for both of RKF and MSKF. The MSKF column filtrate showed slightly increase in the pH values. It is suggested due to the effect of chemical pre-treatment using NaOH solution.

3.3 TDS removal by RKF

The effect of flow rate on the TDS of filtrates of the RKF adsorbent column can be seen in Fig. 2. Fig. 2 reveals the

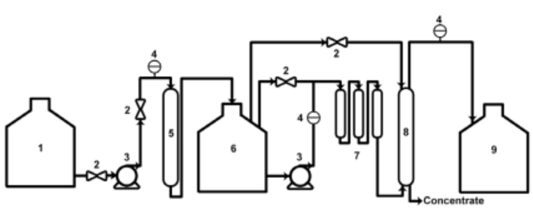


Fig. 1 Schematic of PW treatment; 1. Feed tank; 2. Valve; 3. Centrifugal pump; 4. Pressure gauge; 5. Kapok fiber column; 6. Tank; 7. Sediment filter; 8. Ultrafiltration membrane; 9. Permeate tank

Table 1 Characterization of produced water

Parameter	Value	Standard	Unit
Temperature	21.9	40	°C
pH	7.7	6–9	–
TDS	12,700	4000	mg/L
Phenol	9.9	0.5	mg/L
Barium	5.1	2	mg/L

decrease in TDS of PW using RKF followed by UF membrane. At a flow rate of 5 L/min. the highest TDS removal by RKF is 51.8%. As seen, the TDS removal of UF membrane permeate also increased with increasing the TMP and flow rate. However, an increased in flow rate will increase the compactness of the KF bed in the adsorption column, which implies reducing the TDS removal. From the results, it can be concluded that using either RKF nor combining RKF and UF cannot reduce the TDS to the value of standard regulation. This is because of adsorption process using RKF hindered by surface wax and lignin. Therefore, chemical treatments such as alkali treatments and acid treatments are used to remove pectin, lignin, natural oils, and wax that envelope the outer surface of RKF. The successful treatment of KF will change their properties from hydrophobic to hydrophilic [16].

3.4 TDS removal by MSKF

The effect of contact time on TDS removal in the MSKF adsorption column and UF membrane can be seen in Fig. 3. Using MSKF at varied flow rate and TMP show that the TDS values were in the range of 5395–5890 mg/L. There is an increase in TDS removal compared to those RKF. It is proved that delignification using NaOH solution effectively removes lignin in the surface of Kapok fiber and increases their hydrophilicity, thus increasing the TDS adsorption.

Nevertheless, the TDS values still exceed the regulation standard. Further treatment using UF membrane, the TDS values were between 3410–3570 mg/L. This value meets the regulation standard requirement (4000 mg/L).

As seen in the Fig. 3, TDS values of MSKF filtrate at different flow rate and TMP were between 5395 to 5850 mg/L. However, TDS values of the UF permeates were less than 4000 mg/L and met the standard. The decreased of TDS using combined RKF and UF are between 570–700 mg/L while for combined MSKF and UF are in the ranges of 1985–2280 mg/L. The differences are occurred because cleaning process of ultrafiltration membrane after we changed adsorbent from RKF to MSKF. Increasing in the flow rate, the MSKF may experience pore blocking by TDS on its surface due to the saturation of the pores/active site because the average pore size of the KF is mainly concentrated on 3–4 nm [14]. It is suggested that buildup of solids on the MSKF also considered as effect of flowrate. Decreasing the flowrate allowed more time for saturation, since the contact time is greater, and breakthrough time was found to be greater which enhanced the adsorption [22].

Table 2 Acidity of filtrate after 90 min. contact time

Flow-rate (L/min)	Time (min)	RKF	MSKF	TMP (bar)	RKF + UF	MSKF + UF
	30	7.84	7.70	0.25	7.65	7.69
	60	7.70	7.58		7.70	7.88
	90	7.67	7.76		7.81	7.92
	30	7.64	7.75	0.35	7.68	7.95
5	60	7.67	7.83		7.64	8.13
	90	7.62	7.90		7.80	8.04
	30	7.65	7.96	0.50	7.75	7.75
	60	7.52	7.79		7.84	8.07
	90	7.58	7.82		7.79	7.80
	30	7.68	7.83	0.25	8.03	7.89
	60	7.73	7.82		7.91	7.93
	90	7.77	7.78		7.82	8.18
	30	7.69	7.77	0.35	7.79	7.95
6	60	7.61	7.69		7.66	7.97
	90	7.60	7.75		7.68	8.15
	30	7.59	7.79	0.50	7.64	7.93
	60	7.62	7.84		7.63	7.97
	90	7.59	7.82		7.60	8.08
	30	7.43	7.83	0.25	7.85	7.93
	60	7.70	7.80		7.96	7.96
	90	7.75	7.89		7.90	8.00
	30	7.76	7.81	0.35	7.84	7.91
7	60	7.84	7.78		7.89	7.95
	90	7.73	7.69		7.74	8.00
	30	7.60	7.71	0.50	7.68	7.89
	60	7.75	7.69		7.72	8.00
	90	7.82	7.71		7.78	8.10

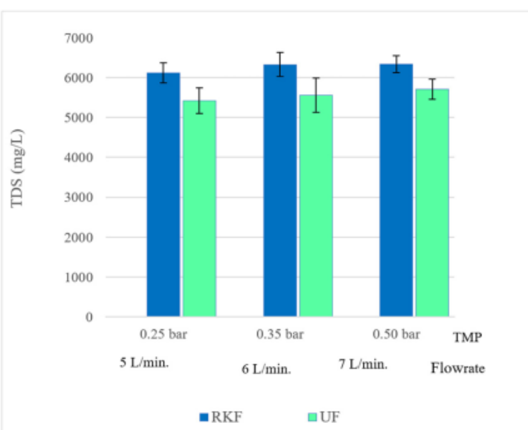


Fig. 2 TDS removal by RKF after 90 min of contact time

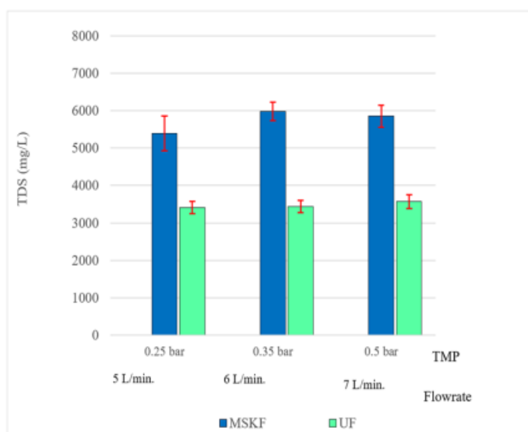


Fig. 3 TDS removal using MSKF after 90 min of contact time

3.5 Phenol removal

Phenol removal using RKF and MSKF is illustrated in Fig. 4. As seen, that at a flow rate of 5 L/min and a TMP of 0.25 bar, RKF removed 62.7% of phenol, followed by 79.1% by the UF membrane. Moreover, at a flow rate of 7 L/min and a TMP of 0.5 bar, RKF and UF removed 52.7% and 60.9% phenol, respectively. At the same condition, the MSKF and combined MSKF-UF remove 72.6% and 84.2% of phenolic compounds. Increases in flow rate and TMP will decrease in phenol removal because using a lower flow rate and a prolong contact time can cause blockage of the membrane pores due to the adsorption process.

Therefore, increasing the contact time will increase the adsorption capacity of an adsorbent until it reaches equilibrium. However, the compactness of the KF bed in the absorbent column needs more attention because the flow rate of filtrates from the adsorbent column will decrease with increases in contact time.

3.6 Barium removal

Barium concentration in PW from oil reservoir are ranges from 0 to 850 mg/L [7] and may reach 1000 mg/L in gas drilling industries [23]. In this work, barium concentration in the PW sample is about 5 mg/L. However, this value still exceeds the regulation. Fig. 5 illustrated that at a flow rate of 5 L/min and a TMP of 0.25 bar, RKF reduced barium concentration by 54.2% followed by UF at 65.8%. An increase in flow rate to 7 L/min and TMP of 0.50 bar; RKF removed 47.3% of barium followed by UF 56.2%. The barium concentration decreases when the flow rate increases due to the release of ions, including barium from the kapok microfibrils.

The MSKF reduced barium from PW in the range of 66.7% at TMP 02.5 bar and 51.1% at TMP 0.5 bar, while

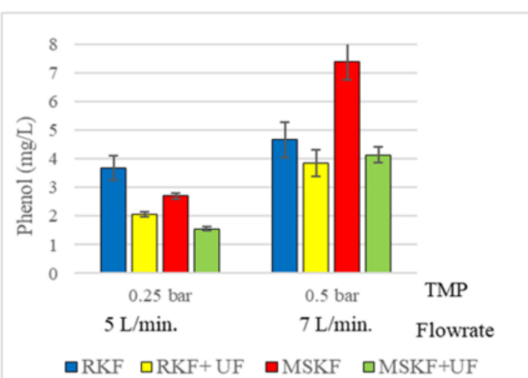


Fig. 4 Phenol removal

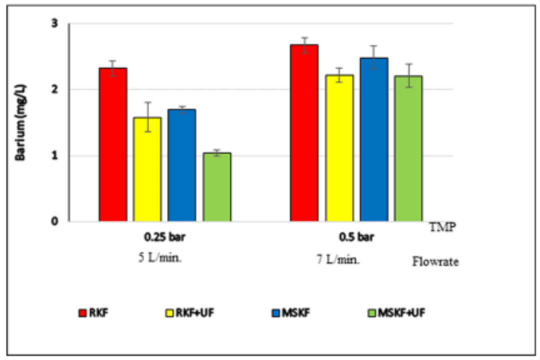


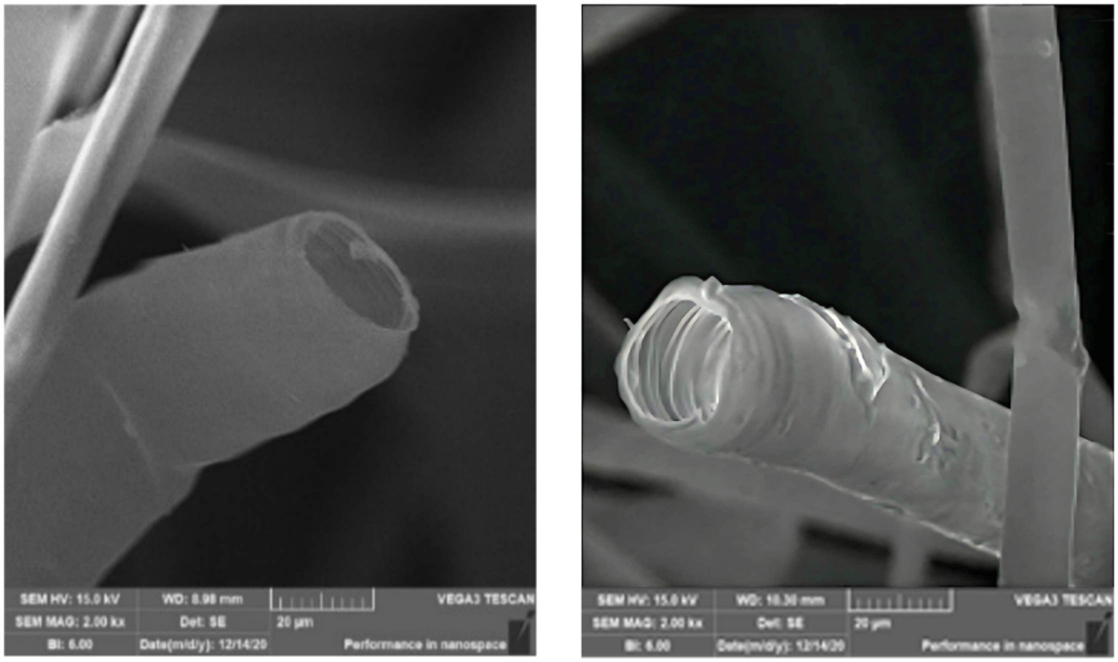
Fig. 5 Barium removal

the combined MSKF and UF decreased barium from 64.0% at 0.25 bar and 56.5% at 0.5 bar, respectively. It seen that MSKF can be use as pretreatment for ultrafiltration membrane in treating the PW. The mechanism of deposition of heavy metal on the surface of Kapok fiber is due to the lumen capillary activity and diffusion [19, 23]. Barium and other metal ions deposition on ultrafiltration membrane may occur via solution diffusion mechanism and supporting by the hydrophilicity of the MSKF. Overall, the average total barium removal of PW in the current experiment is 58.9% and meets the requirements of PW reinjection into reservoirs. The low concentration of barium in the PW sample suggested that barium is origin from barium sulfate (barite), which is used in the oil and gas drilling process as a lubricant. In petrochemical and gas drilling, barite increases the density of all types of mud and prevents damage to the oil drilling equipment.

3.7 SEM-EDX of Kapok fiber

A scanning electron microscope with Energy Dispersive X-ray (SEM-EDX) was used to investigate the surface morphology and identify the elements of KF. Fig. 6 shows the SEM-EDX images of RKF and MSKF after use as an absorbent.

The SEM images in Fig. 6 (a) show that the RKF has the smooth outer surface. The RKF surface smoothness can be attributed to the wax that is adhered on the fiber [24]. In contrast, in Fig. 6 (b), the MSKF surface looks rough, cracked, with narrow lumen. The rougher RKF is caused by wax and cuticle loss from the kapok surface and exposure of fibrils to the surface. Hot water increases the fibrous structure of kapok because it removes the volatile compounds, waxy substances, and other impurities from the fiber surface and thus facilities sorption. In addition, the RKF alkaline pre-treatment revealed that the hydroxyl



(a) (b)

Fig. 6 SEM images of (a) RKF, (b) MSKF after adsorption

and methoxy present in the interpenetrating polymer network that can adsorb heavy metal ions because of its fishnet structure [20]. Therefore, it increases the adsorption capacity of the sorbent. It is found that RKF becomes compact and shrinks after being used as adsorbent.

Fig. 7 shows the EDX spectra of RKF, RKF after adsorption, and MSKF after use as adsorbent. The rough surface exhibits wax loss from RKF and subsequent exposure to hydrophilic hydroxyl groups, which are advantageous in fiber-matrix adhesion as they facilitate interlocking reactions and mechanical binding to chemicals such as resins and dyes. The coarse RKF is caused by the removal of wax and cuticle from the surface structure and exposure the fibrils to hot water. Therefore, pretreatment of RKF using NaOH and hot water changes the fiber surface topography [13].

Fig. 7 (a) shows that the primary RKF elements is carbon, oxygen and aluminum. Fig. 7 (b) and (c) show the RKF and MSKF after being used for the sorption of PW containing barium, chlorine, carbon, oxygen, aluminum, and sodium. There is an increase in barium from 0.02 % using RKF to 0.12% when using MSKF. It is also shown that the PW contains sodium chloride, one of the compounds which characterized the PW as saline water. The MSKF could be a fibrous natural material sorbent type that can adsorb barium and other ions, primarily sodium and chlorine from PW. Therefore, it can be used to decrease the salinity of PW.

4 Conclusions

The modified surface of Kapok fiber using 5% NaOH and hot water at 98.5 °C change the surface topography of Kapok fiber to be coarser and hydrophilic, thereby increasing the adsorption of metal ions such as aluminum, sodium, and barium from produced water. The removals of TDS, phenol, and barium by MSKF were 57.5%, 72.6%, and 66.7% respectively, at a flow rate of 5 L/min and contact time of 90 min. The optimum percentage of decrease in TDS, phenol, and barium by the combined MSKF and ultrafiltration membrane was 94.3%, 84.2%, and 64%, respectively at a TMP of 0.25 bar.

The MSKF is the potential for TDS, phenol and barium removal. The modified surface of Kapok fiber using chemicals changes its hydrophobic properties, thus increasing its hydrophilicity. Using a high flow rate and prolong time can cause blockage of the Kapok fiber pores due to the filtration process. The filtrates/permeates obtained has met

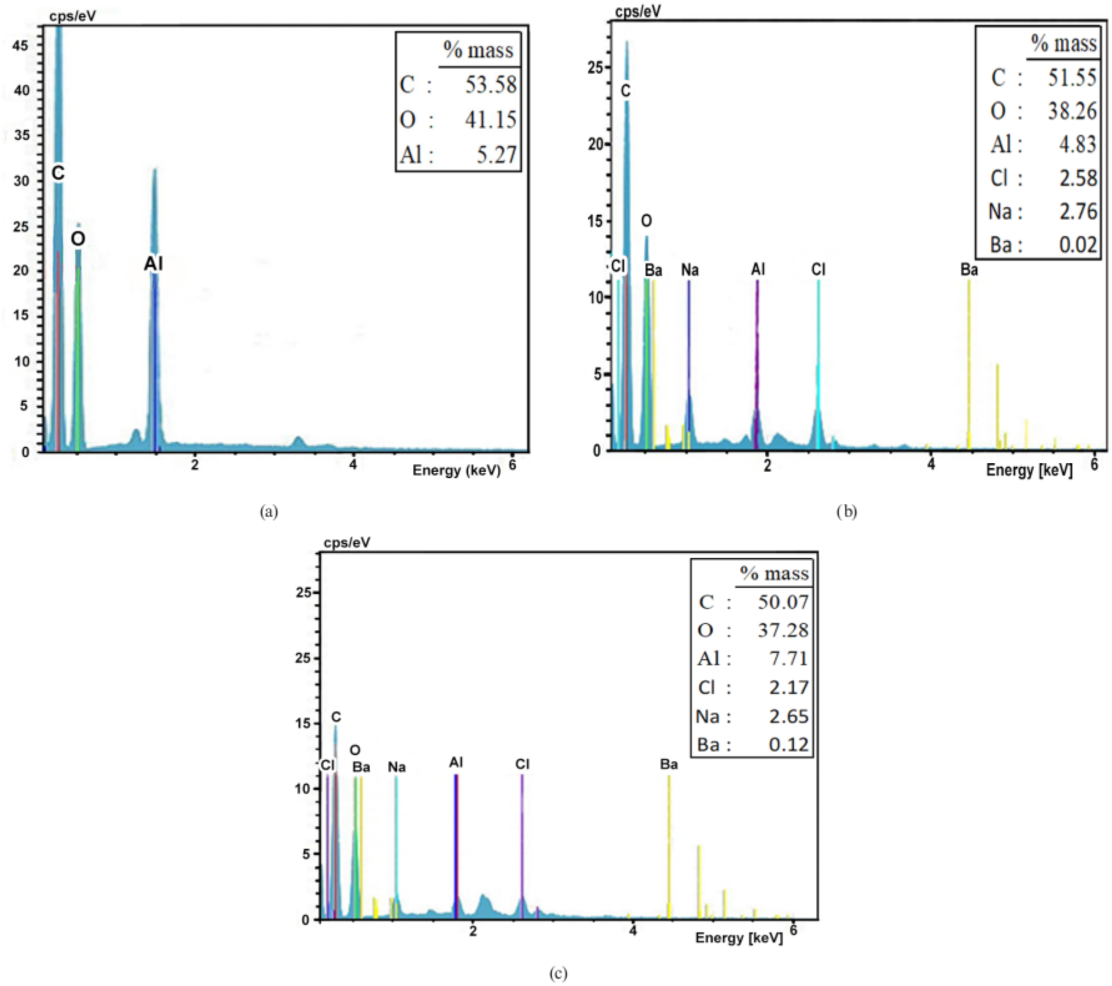


Fig. 7 (a) EDX spectra of RKF; (b) EDX spectra of RKF after adsorption; (c) EDX spectra of MSKF after adsorption

the conditional water in wastewater quality stipulated by Indonesian standards. Therefore, the modified surface of Kapok fiber potentially used not only for oil absorbent but also adsorption of metal ions from produced water.

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