Removal of dimethoate from contaminated water using corncob and rice husk as low-cost materials

Sakine Ugurlu Karaağaç*, Ahmed Mohamed Ramdan Abudaıa

Department of Environmental Engineering, Faculty of Engineering, Karabuk University, Karabuk 78050, Turkey,
emails: sakineugurlu@karabuk.edu.tr (S.U. Karaağaç) ORCID Number: 0000-0003-4673-706X,
ahmedbudya851@gmail.com (A.M. Ramdan Abudaıa) ORCID Number: 0000-0002-2571-7185

ABSTRACT

This research aims to evaluate the potential removal efficiency of dimethoate from aqueous solutions utilizing activated carbon (AC) derived from corncob and rice husk. First, researchers identified the structural and morphological features of produced corncob activated carbon (KOHCC) and rice husk activated carbon (KOHRH). To examine proximate analyses and characterize changes on the surfaces and structure of corncob following chemical activation with KOH, X-ray diffraction, scanning electron microscopy, and energy-dispersive X-ray spectroscopy were utilized. Second, the effects of sorbet dose, sorbet beginning concentration, and contact duration on its usage were explored. Various sorption properties were investigated and compared to commercial activated carbon generated from bituminous coal. According to the findings, batch adsorption was carried out using dimethoate solutions ranging from 10 to 150 mg/L. The maximum removal percentage of dimethoate by KOHRH was 93.1% for 10 ppb and 93.9% for 100 ppb at 0.5 mg/100 mL. In contrast, the most significant removal effectiveness of dimethoate by KOHCC was 88.7% for 10 ppb at 0.4 mg/100 mL respectively. Therefore, dimethoate may be removed from water using KOHCC and KOHRH.

Keywords: Activated carbon; Corncob; Dimethoate removal; Rice husk; Water pollution

1. Introduction

It is inevitable to use agricultural pesticides to eliminate agricultural pests, enhance agricultural production, and improve product quality. However, on the other hand, there is no doubt that the excessive use of these agricultural pesticides has a direct negative impact on the environment and human health, from diarrhea to death [1]. Water contamination caused by pesticides used in agriculture and public health is a severe problem worldwide [2]. When agricultural pesticides are applied, they penetrate the soil, contaminating water bodies by direct runoff, leaching, and percolation [3]. Pesticides (hazardous substances) are commonly used in agricultural activities [4]. Pesticides, dyes, heavy metal ions, etc., are responsible for contaminating the water. These toxic substances destroy the water quality and make it unsuitable for irrigation and domestic purposes [5].

Furthermore, have inevitable negative consequences such as toxicity, carcinogenicity, and mutagenicity on the environment. Pesticides in water can create significant environmental and human health issues [6,7]. Pesticides are harmful materials discovered in surface water bodies due to agricultural runoff. Pesticides' negative impact on human health and the environment has led to rigorous drinking water quality regulations in several nations [7,8]. Pesticide usage has aroused considerable concerns in this setting, not just about possible effects on human health but also about effects on animals and fragile ecosystems [9–11]. As a result, maintaining and improving water quality is vital in

* Corresponding author.

living, and the need is constantly expanding, highlighting the need for effective remedial strategies [6,7].

Agricultural runoff might be one of the primary sources of water pollution that is difficult to eliminate using standard treatment technologies such as oxidation, R.O., ion exchange, electro-dialysis, adsorption, and so on [12–15]. According to Ahmad et al. [3], Adsorption is the best wastewater treatment technique due to its universality, cheap cost, and ease of operation. Organic contaminants, both soluble and insoluble, can be removed by adsorption. This approach has a removal capability of up to 99.9%. Adsorption has been utilized to remove a range of organic contaminants from various polluted water sources due to these findings [10].

Carbonaceous materials, as they are known, held a distinctive position among the main adsorbents for a long time to be capable of adsorbing diverse organic molecules. Because of its large surface area and porosity, activated carbon is particularly effective in removing many pesticides from water and wastewater. It is a versatile material that can be used in a variety of technical operations. Granular activated carbon (GAC) [16], powdered activated carbon (PAC) [17], and commercial activated carbon (CAC) have all been employed in this study [18]. GAC and PAC forms are the most commonly utilized because they are considered extremely capable and practical materials for pesticide adsorption [19].

As a result, several researchers have attempted to synthesize activated carbons to remove different contaminants utilizing renewable and less expensive precursors, primarily industrial and agricultural leftovers in recent years. Carbons have been synthesized from a wide range of biomass and other wastes, including dates stone [20], wood [21], and oil palm fronds [22].

This research aims to evaluate the potential efficiency of activated carbon (AC) produced from corncob and rice husk to remove dimethoate from aqueous media (solution) efficiently. In this case, the structural and morphological parameters of produced corncob activated carbon (KOHCC) and rice husk activated carbon (KOHRH) were determined. Second, various relevant sorption factors were compared to commercial activated carbon (CAC) derived from bituminous coal.

2. Materials and methods

2.1. Activated carbon from corncob and rise husk

Corncob was collected from local corn growers (Zeamais) in Karabuk, Turkey, while rice husk was obtained from a rice producer in Edirne, Turkey. Physical activation was used to create corncob and rose husk-activated carbon. The raw materials were chopped into 1–3 cm pieces and dried in a 105°C oven for 24 h [23]. Raw materials were carbonized at 450°C for 1.5 h under a nitrogen gas flow. Under these oxygen-deficient circumstances, corncobs were thermally decomposed to porous carbonaceous materials and hydrocarbon molecules. The chars were removed, crushed, and sieved to a uniform size of 1.0–1.5 mm.

Carbonized corncob and rice husk were soaked in an immersed KOH proportion ratio of 1:1 (KOH: corncob/rice husk) in a stainless beaker for 12 h before being dried at 105°C in an oven overnight. To commence activation,

temperature was increased at 780°C for 1 h with a flow of N_2 3 dm^3/min. The resulting activated carbon was reduced to 250°C before being treated with deionized water [6].

To remove the residual ash content, the samples were steeped in 0.1 M hydrochloric acid (HCl) for 1 h. They were then rinsed with hot distilled water until the pH reached 6–7, dried overnight at 105°C, and labeled KOHCC activated carbon for corncob and KOHRH activated carbon for rice husk. In contrast to the two local activated carbons, commercial activated carbon (AC) made from bituminous coal was used as a reference KOHCC and KOHRH) [24].

2.2. Pesticides

Dimethoate is a common agricultural pesticide that can build in significant quantities in surface water sources. Dimethoate (99.5%) was obtained from Reference Chemistry Company Ankara, Turkey, whereas ISOLAB Chemical Company in Turkey provided hydrochloric acid (analytical grade), liquid chromatography gradient grade methanol, and acetonitrile. To prepare a stock solution with a concentration of 1 mg/mL dimethoate, a 10 mg of dimethoate was dissolved in 10 mL methanol, and a separate working standard was made by diluting with distilled water; all solutions were prepared using distilled water. Table 1 depicts the pesticide formulations and some of their properties in this study.
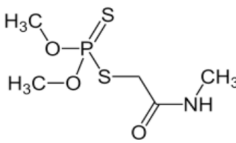
2.3. Specific characterization of corncob and rise husk

The spectrum of the obtained KOHCC and KOHRH activated carbon was determined at a wavelength ($\lambda = 0.15406$ nm) using X-ray diffraction (XRD) on Ultima IV, Rigaku, Japan, with graphite monochromatized CuKa and the 1.0 s^{-1} scanning rate with a 0.02° scanning step applied in recording the pattern at 10°–90° range of Cu radiation with Ni filter which was given at 40 kV and 20 mA, as shown in Fig. 1a and b, the steep peaks are located at 23.3° and 24.4°, respectively. Scanning electron microscopy (SEM) of the KOHCC and KOHRH were taken using the JOEL JSM-6610LV, Japan, and compositional analysis was performed using energy-dispersive X-ray spectroscopy (EDAX).

2.4. Batch adsorption studies

To evaluate the pesticide adsorption isotherms, activated carbons were used in this study; batch experiments were used as the basis [25]. Adsorption studies were carried out

Table 1
Physical–chemical properties of dimethoate used in this study

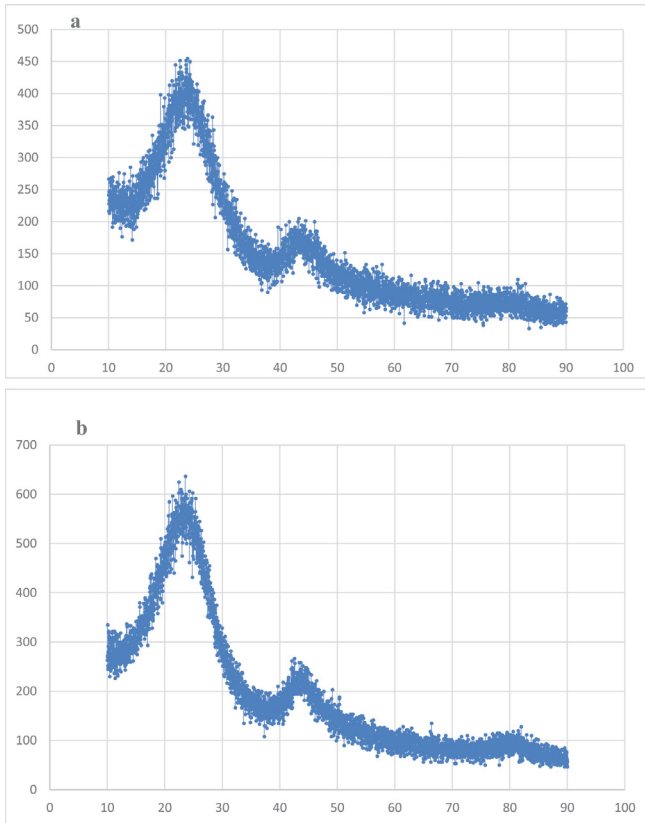| Structure	
Type	Organophosphate
Molecular weight (g/mol)	229.26 g/mol
Solubility in water at 20°C (mg/L)	2.5 g/100 mL
$\log K_{ow}$	0.78

Fig. 1. XRD patterns of KOHCC (a) and KOHRH (b).

in 1 L containers containing 1 mg/mL dimethoate as a stock solution. Dimethoate stock solution was serially diluted with distilled water to achieve various concentrations. In the sciences of kinetics and equilibrium. In 500 mL Erlenmeyer flasks, 250 mL of dimethoate at beginning concentrations of 20–100 mg/L were agitated with appropriate dosages of KOHCC and KOHRH. The mixture solution was shaken at 150 rpm for 90 min at room temperature, and samples were taken at different suitable intervals (5, 10, 25, 30, 45, 75, 90 min, and 24 h). The mixture in the vial was filtered via a 0.45 m film tube linked to a syringe to reduce carbon fines interference with the analysis. The concentration of dimethoate was measured using LCMS Agilent with a detector named UV/VIS SPD-20A. An Inertsil ODS-4, C18 reverse phase analytical column (4.6 and 250 mm length, 5 m particle size; GL Sciences, Japan) was utilized for separation. Binary gradient chromatography was performed at a flow rate of 1.0 mL/min with an optimum acetonitrile-to-water mixture ratio as the mobile phase. A UV detector with a wavelength of 210 nm was used at a specific retention time. The quantity of dimethoate sorbed at time t, q_t (mg/g) and equilibrium, q_e (mg/g) and the removal efficiency of adsorption (%) onto KOHCC and KOHCC) were calculated using Eqs. (1)–(3).

$$q_t = \frac{(C_0 - C_t)V}{W} \tag{1}$$

$$q_e = \frac{(C_0 - C_e)V}{W} \tag{2}$$

$$\text{Removal efficiency}(\%) = \frac{(C_0 - C_t)}{W} \times 100 \tag{3}$$

where C_0, C_t, and C_e (mg/L) are the concentrations of dimethoate in the aqueous solution at the starting, time t, and equilibrium, respectively. The volume of the aqueous solution used in the adsorption experiment is V (L), and the mass of the KOHCC and KOHCC used is W (g).

3. Results and discussions

3.1. Surface characterization of prepared adsorbent

The pore size and structure of activated carbon from corncob and rice husk were investigated using SEM. Photographs of the tested activated carbon were taken using a scanning electron microscope. The structure of activated carbons may change due to differences in raw materials utilized in manufacturing. Figs. 2–5 show SEM images of two activated carbons. KOHCC's SEM micrograph (Fig. 2) exhibited rough spots with long ridges. The morphology of a highly porous, spongy, and branched particle is depicted in this image. The anticipated spongy and porous structure may also be seen in the KOHRH SEM image (Fig. 3). The SEM images of KOHRH and KOHCC show that the ACCS surface has well-developed pores. On the surface, there were several huge pores in the honeycomb shape. The SEM photos demonstrate that both activated carbons had many well-developed holes on the surface, indicating the likelihood of pesticide adsorption.

The EDAX spectra reveals significant peaks for corncob Table 2 as C, P, O, Cl, and Fe, with average percent values of 73.4%, 18.06%, 5.10%, 1.65%, 4.08%, and 0.72%, respectively.

KOHCC's SEM microscopy shows an irregular pore structure with varying diameters and cavities.

The EDAX spectra reveal significant peaks for rice husk (Fig. 6) Table 3 as C, P, O, Si, and Fe, with average percent values of 66.78%, 18.21%, 5.53%, 4.18%, and 4.08%, respectively.

The EDAX spectrum shows firm peaks for rice husk Table 3 as C, P, O, Si, and Fe with average percent of 66.78%, 18.21%, 5.53%, 4.18%, and 4.08%, respectively.

3.2. Removal efficiency

Table 4 shows the removal effectiveness of dimethoate using various activated carbons at 0.1–0.6 mg/100 mL in produced pesticide solution with different beginning concentrations of dimethoate (10 ppb and 100 ppb).

As illustrated in Fig. 7, the maximum removal percentage of dimethoate by KOHRH was 93.1% for 10 ppb and 93.9% for 100 ppb at 0.5 mg/100 mL. As shown in Fig. 8, the most significant removal effectiveness of dimethoate by KOHCC was 88.7% at 10 ppb and 85.1% at 100 ppb at 0.4 and 0.2 mg/100 mL, respectively. On the other hand, commercial activated carbon (AC) had the highest removal rate of approximately 95.0% for both concentrations. Nonetheless, the data showed that the two local activated carbons had an excellent adsorption capacity for dimethoate; they were determined to have an adsorption capacity nearly comparable to commercial activated carbon. As a result, two local activated carbons might be used in place of commercial
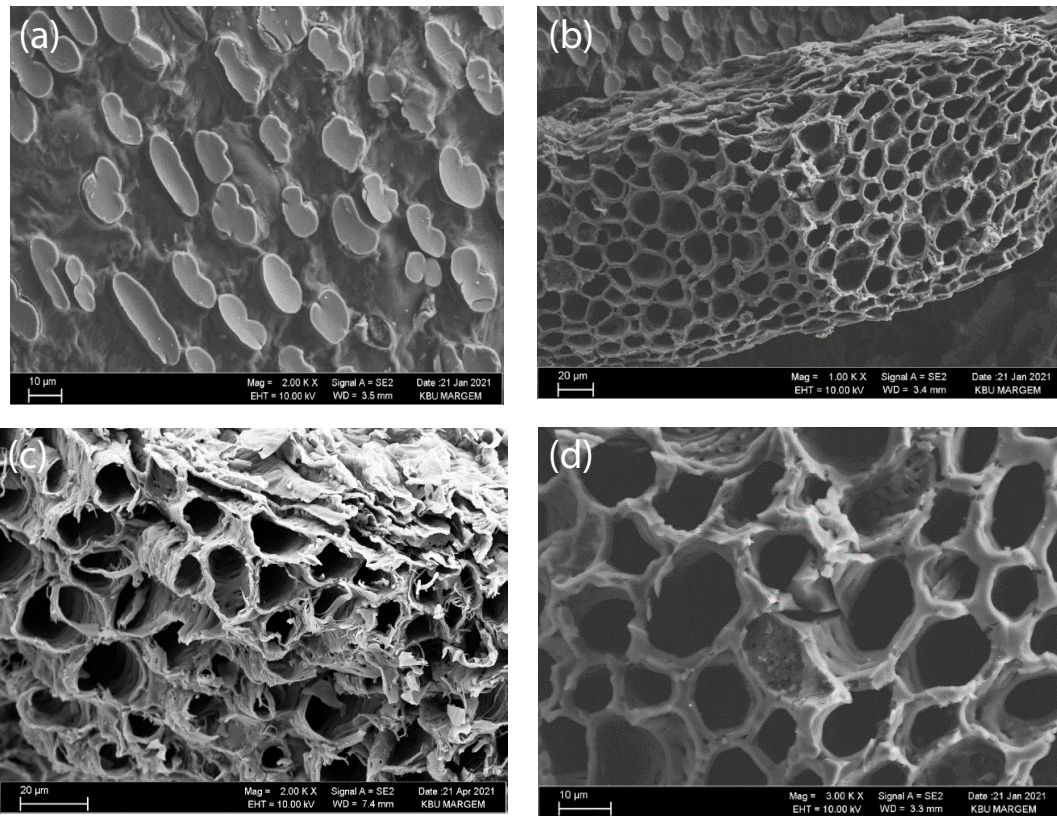
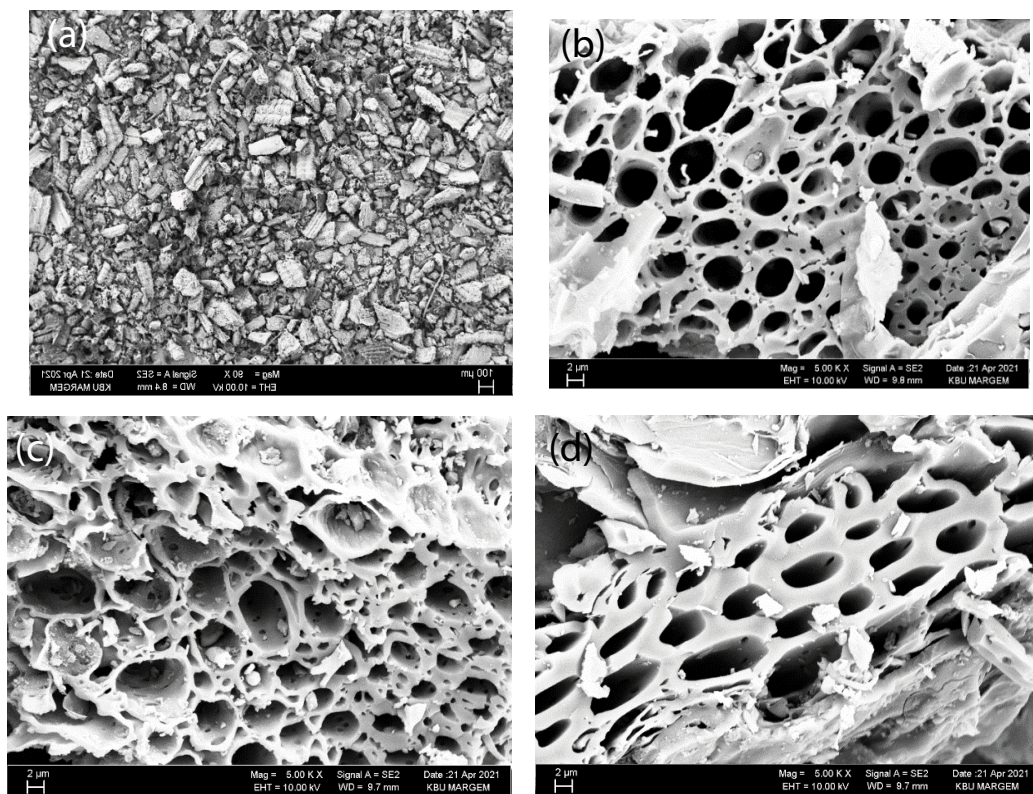Fig. 2. SEM micrograph of activated carbon from corncob (KOHCC).

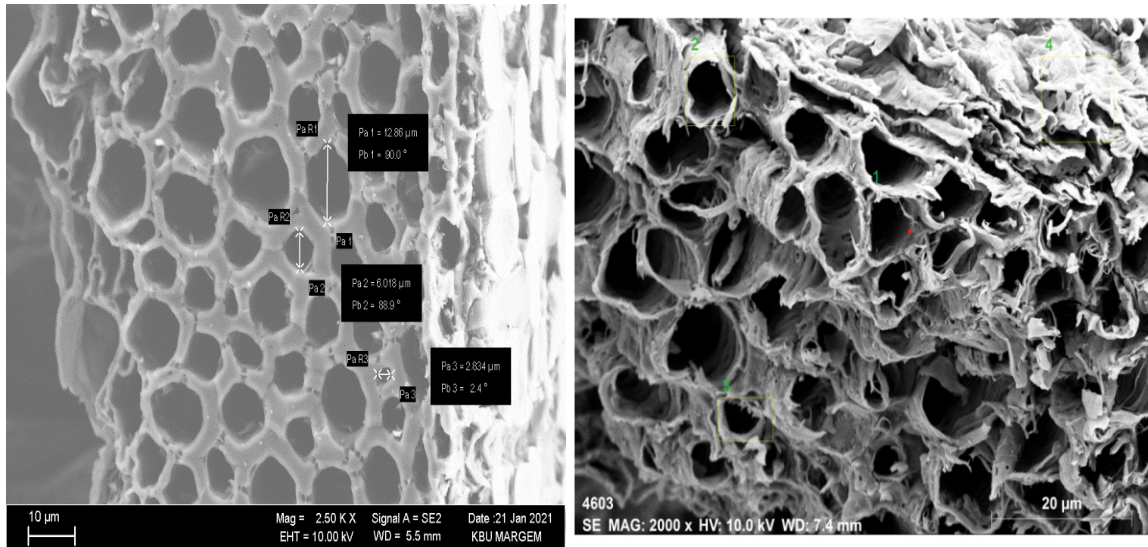Fig. 3. SEM micrograph of activated carbon from rice husk KOHRH.

Fig. 4. Pore size of corncob activated carbon.

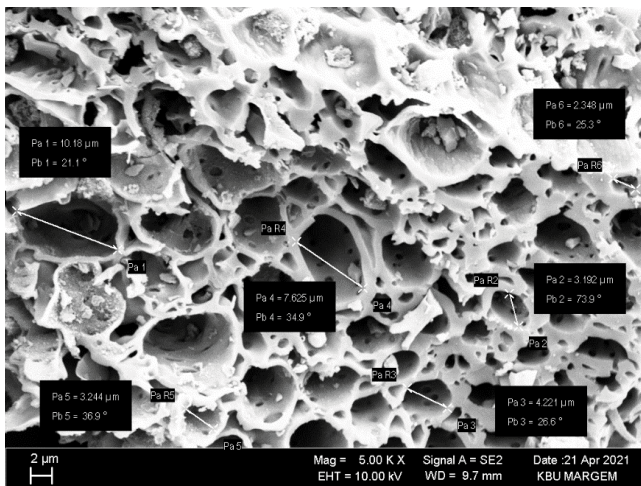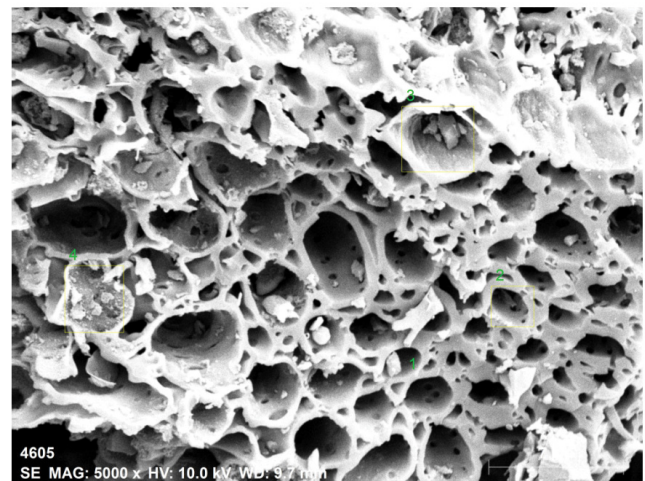Fig. 5. Pore size of rice husk activated carbon.

Fig. 6. Rice husk (KOHRH) EDAX spectra after activation.

Table 2
Corncob (KOHCC) EDAX spectra after activation

Mass % spectrum	C	N	O	Si	P	S	Cl	K	Ca	Fe	Cu	Se
1	78.13	0.00	7.41	0.00	13.15	0.19	0.81	0.00	0.31	0.00	0.00	0.00
2	83.57	0.00	2.85	0.25	11.45	0.00	0.38	0.37	0.07	1.06	0.00	0.00
3	54.02	0.00	0.07	0.32	38.08	0.00	5.02	2.22	0.00	0.00	0.00	0.26
4	77.90	0.00	10.08	0.01	9.57	0.00	0.38	0.18	0.07	1.82	0.00	0.00
Mean value	73.41	0.00	5.10	0.014	18.06	0.05	1.65	0.69	0.11	0.72	0.00	0.07
Sigma	13.18	0.00	4.49	0.16	13.43	0.10	2.26	1.03	0.14	0.89	0.00	0.13
Sigma mean	6.59	0.00	2.24	0.08	6.71	0.05	1.13	0.52	0.07	0.44	0.00	0.07

activated carbon as low-cost–high-performance adsorbents for dimethoate elimination. KOHRH, for example, has a larger adsorption capacity than KOHCC and is approximately as effective as AC.

3.3. Effect of contact time and initial concentration

The equilibrium contact time is essential in deciding adsorbent adsorption capacity. Figs. 9 and 10 show the effects of contact time and initial concentration on dimethoate

Table 3
EDAX spectra of corncob (KOHRH) after activation

Mass % spectrum	C	N	O	Si	P	S	Cl	K	Ca	Fe	Cu	Se
1	61.94	0.00	5.09	3.89	25.14	0.00	1.62	0.43	0.00	1.89	0.00	0.00
2	69.64	0.00	1.14	0.69	18.70	0.16	0.42	0.00	0.09	9.17	0.00	0.00
3	79.79	0.00	2.38	1.41	15.14	0.00	0.63	0.12	0.26	0.00	0.11	0.16
4	55.77	0.00	13.54	10.72	13.87	0.20	0.13	0.10	0.34	5.25	0.00	0.09
Mean value	66.78	0.00	5.53	4.18	18.21	0.09	0.70	0.16	0.17	4.08	0.03	0.06
Sigma	10.36	0.00	5.58	4.57	5.05	0.10	0.64	0.19	0.16	4.03	0.05	0.08
Sigma mean	5.18	0.00	2.79	2.28	2.53	0.05	0.32	0.09	0.08	2.01	0.03	0.04

Table 4
Removal efficiency of dimethoate using 0.1–0.6 mg/100 mL selected activated carbons in sample solution

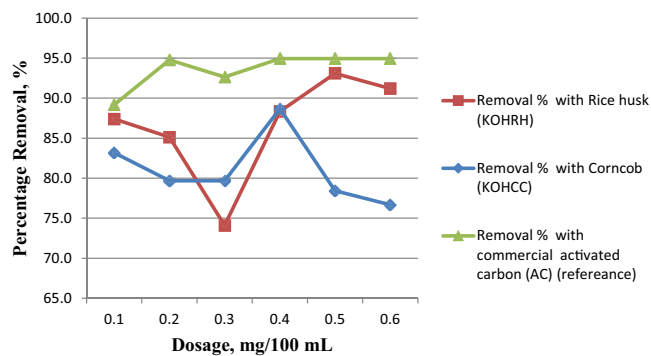|	Dosage mg/100 mL	Removal % (KOHRH)	Removal % (KOHCC)	Removal % (AC) reference
10 ppb (10.000) dimethoate solution	0.1	85.1	79.7	94.8
	0.2	74.1	79.7	92.6
	0.3	88.4	88.7	94.9
	0.4	93.1	78.4	94.9
	0.5	91.2	76.7	94.9
	0.6	87.2	82.8	85.5
100 ppb (100.000) dimethoate solution	0.1	84.8	85.1	94.7
	0.2	89.3	82.0	95.0
	0.3	90.0	82.7	94.9
	0.4	93.9	81.8	94.8
	0.5	92.0	79.6	94.8
	0.6	85.1	79.7	94.8

Fig. 7. Removal efficiency of dimethoate using different dosages of KOHRH, KOHCC, and AC as reference in pesticide solution with 10 ppb concentration.
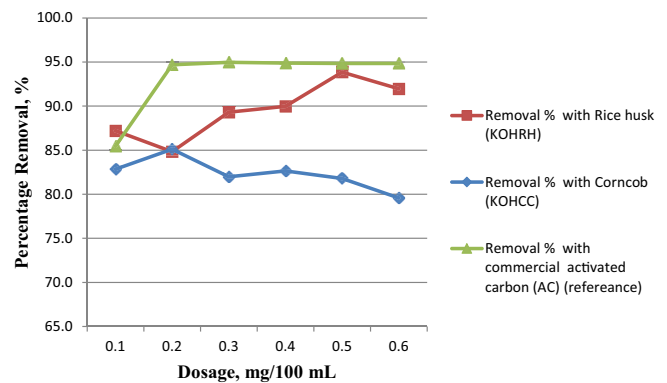

Fig. 8. Removal efficiency of dimethoate using the different dosages of KOHRH, KOHCC, and AC as reference in pesticide solution with 100 ppb concentration.

removal by KOHRH, KOHCC, and AC. The adsorption and starting concentrations were investigated at various contact times (5, 10, 25, 30, 45, 75, 90, and 120 min). For this analysis, two starting concentrations of 10 and 100 ppb were used, while the adsorbent dosage remained constant with the ideal from the previous section at a temperature of 30°C.

Dimethoate absorption was reported to be quick for the first 10 min before progressively slowing down until equilibrium was reached. After the 75 min equilibrium period, no significant adsorption was detected; nevertheless, the removal efficiency of dimethoate from stock solution by KOHRH is 93.1% and 93.9% for 10 and 100 ppb, respectively, during the 75 min contact time.

Figs. 9 and 10 depict the influence of contact time on pesticide removal efficiency. Adsorption is often divided into three stages: (1) instantaneous adsorption, (2) sequential gradual adsorption, and (3) equilibrium [26]. Adsorption increased with increasing duration for the pesticide combination investigated, and it was quick in the beginning before
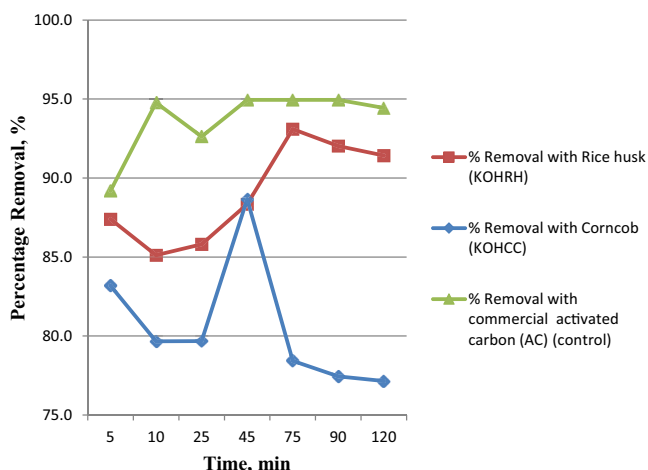
Fig. 9. Influence of contact time on the removal of 10 ppb dimethoate initial concentration using activated carbon.
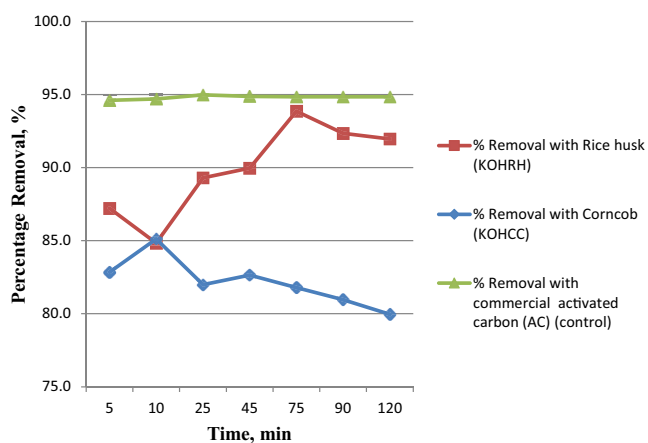

Fig. 10. Influence of contact time on the removal of 100 ppb dimethoate initial concentration using activated carbon.

slowing down until equilibrium was reached after 75 min. An increase in contact duration had a minor influence on pesticide combination elimination. Pesticide adsorption followed a similar pattern [23].

As demonstrated in Figs. 9 and 10, the removal efficiency of dimethoate from stock solution by KOHCC is 88.7% for 10 ppb and 85.1% for 100 ppb at 45 and 10 min of contacting time, respectively. In the trials, activated carbon achieved the highest removal rate in the starting solution of 10 ppb in 45 min and the starting solution of 100 ppb in 25 min. As a result, two local activated carbons might be used in place of commercial activated carbon as low-cost–high-performance adsorbents for dimethoate elimination. KOHRH, for example, has a larger adsorption capacity than KOHCC and is approximately as effective as AC.

4. Conclusion

Pesticides have been demonstrated to adsorb on the surface of activated carbon significantly. Because of the interaction between the chemicals in river water and the surface areas of the adsorbents, activated carbons formed from waste products were investigated as prospective adsorbents for drinking water protection in this study. The degree of pesticide removal rose as the initial pesticide concentration decreased and the number of adsorbents utilized increased; it was also affected by the pesticides' hydrophobic qualities. According to the findings, batch adsorption was performed using dimethoate solutions ranging from 10 to 150 mg/L. At 0.5 mg/100 mL, the highest removal percentage of dimethoate by KOHRH was 93.1% for 10 ppb and 93.9% for 100 ppb. On the other hand, dimethoate removal efficacy by KOHCC was 88.7% for 10 ppb at 0.4 mg/100 mL, respectively.

Two local adsorbents were compared to commercial activated carbon in adsorption capacity. According to the findings of this study, activated carbon derived from corncob (KOHCC) and rice husk (KOHRH) might be employed as a low-cost, high-performance adsorbent alternative to commercial activated carbon in wastewater treatment for pesticide removal.

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