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Adsorption performance of human-like collagen by alkali-modified Kapok fiber: a kinetic, equilibrium, and mechanistic investigation

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Abstract: Collagen-based dressings achieve excellent repair of the skin during medical cosmetology, which has received a lot of attention recently. Although great progress has been made on using biomass fiber as dressing carrier, more research is required on developing novel biomass fibers because of the limitations of (i.e., high cost and complex processing) of existing materials. In this study, the adsorption behaviors of two human-like collagen were studied by examining the Kapok fiber that was modified using alkali consisting of various amounts of the mass fraction. Results show that the alkali-modified Kapok fiber surface becomes rough with vertically arranged grooves, and the cross-section depicts the hollow cavity structure. The composition analysis of alkali modified Kapok shows that alkali dissolves part of the hemicellulose and lignin. Additionally, the surface energy rises sharply and the water contact angle changed from hydrophobic to hydrophilic. The adsorption amount of raw Kapok fiber is around 0.6g/g, which accounts for only one twenty-first of the adsorption amount of alkali-treated Kapok (around 12.6g/g), while the equilibrium adsorption amount was not sensitive to alkali concentration. The kinetics of human-like collagen followed both Quasi first and Quasi second order kinetic model, implying that the adsorption process was characterized by both physisorption and chemisorption. Finally, characterization of the AKF-2 coupled with the studies based on the inter-particle diffusion model showed a three-step of human-like collagen diffusion consisting of surface diffusion, inter-fiber diffusion and fiber' hollow cavity diffusion. Our results demonstrate a perfect high absorption performance of Kapok fiber providing a potential for application of collagen-based dressings.

Keyword: Alkali-modified Kapok; human-like collagen; adsorption kinetics; diffusion;

Introduction

Since the 1970s, medical cosmetology has become one of the fastest-growing and closest-commercialized industries. Medical cosmetology primarily focuses on repairing and reshaping various parts of the human body through drugs, surgery, medical equipment and other traumatic or irreversible

30 medical technology methods. However, the application of medical cosmetology can inevitably
31 damage the barrier function of the skin and cause postoperative complications, including skin
32 pigmentation, contact dermatitis, superficial ulcers and other skin diseases. Among several methods
33 to deal with skin damage, postoperative repair dressing is one of the most widely and commonly used
34 methods. Recently, collagen is the main dressings used in postoperative repair dressings to deal with
35 skin damage. The research of collagen-based dressings are mainly focused on three areas: 1)
36 preparation (Ge et al., 2018); 2) healing mechanism (Ge et al., 2018; Lozeau et al., 2018; Pallaske et
37 al., 2018); and 3) application (Ye et al., 2014). When collagen is used, a biomass fiber based dressing-
38 carrier is widely used including seaweed (Andryukov et al., 2020), chitin (Hao et al., 2020) and
39 protein (Guo et al., 2020) fibers. Although great progress has been made on using biomass fiber as
40 dressing carrier, more research is required on developing novel biomass fibers because of the
41 limitations of existing materials (high cost and complexity of process). Additionally, novel biomass
42 fiber can help to maintain product diversity and provide avenues of opportunities to examine the
43 effects of different biomass fiber dressings on skin problem.

44 Kapok is a biomass fiber known for its lightweight (Yang et al., 2018) and thinness ($20\sim45\ \mu\text{m}$)
45 among all the natural fibers. Single Kapok fiber has a big cylindrical hollow cavity structure and is
46 characterized nearly transparent and very thin cell wall (Abdullah et al., 2010). Moreover, it has a
47 hollow rate of $>86\%$, which is the highest hollow rate in nature fiber (Xiang et al., 2013). It has
48 excellent moisture absorption and breathability characteristics due to the physicochemical property
49 mentioned above (Yang et al., 2018). Furthermore, Kapok fiber has superior biocompatibility,
50 biodegradability and natural antibacterial properties, with an antibacterial rate of $>91.9\%$ (Wang et
51 al., 2015). These unique properties have promoted Kapok fiber as dressing carrier materials for the
52 collagen-based postoperative repair dressing. Because of the hydrophobic characteristics of Kapok
53 fiber, earlier studies were mainly focused on oil removal (Dong et al., 2015b, c; Zhang et al., 2021),
54 wastewater treatment (Sun et al., 2018; Wang and Wang, 2018; Wang and Zheng, 2017) as well as
55 dye adsorption (Li et al., 2015; Zhang et al., 2015). Nevertheless, there have been no more studies to
56 evaluate the performance of Kapok as a potential fiber for collagen-based dressings.

57 In collagen-based dressings, adsorption characteristics play an important role in the overall
58 performance of the postoperative repair. In the study, the adsorption performance of alkali modified

Kapok fiber for various human-like collagen was quantified by evaluating the equilibrium time and amount of adsorption. Meanwhile, the effect of different alkali concentrations on the adsorption performance were also examined during this study. Additionally, the adsorption kinetic model of human-like collagen on the Kapok fiber, including two Quasi (first- and second-order kinetic models) and a Weber-Morris model were discussed. Finally, the adsorption process and mechanism were investigated. The alkali-modified Kapok fiber has potential to be used as an alternative material in collagen-based dressings application.

Material and methods

Alkali-modified Kapok fiber

The Kapok fiber used in this experiment came from Pate County, Java Tengah, Indonesia. The samples (6g) were put into a beaker with different NaOH solution mass fractions (2%, 4%, 6%, 8%), which was then placed in a thermostat water bath (HH-4, ZENROE, China) to maintain a constant temperature around 70°C for alkali-modification. After 6 hours of reaction/treatment, the Kapok fibers were washed with deionized water and dried naturally at room temperature (26°C, 68%) to obtain alkali-modified samples. The obtained samples were marked as AKF-2, AKF-4, AKF-6 and AKF-8, which refers to 2%, 4%, 6% and 8% alkali concentration, respectively.

Characterization of Fibers

Their surface and cross-sectional morphologies were examined using field emission scanning electron microscopy (SU8010, Hitachi, Japan) by exerting an accelerating voltage of 20kV on a sample sputter-coated with gold. Fourier transformed infrared (FTIR) spectrometer (Nicolet6700, Thermo Fisher, USA) was used to record diversification between raw and alkali-modified fibers. X-ray diffraction (XRD) experiments were executed on a diffractometer (D/MAX-2500 PC, Rigaku, Japan) by applying Cu Ka radiation ($\lambda=0.154\text{nm}$, 40kV, 200mA) to separate the crystal phase of the raw and alkali-modified fibers. Shredded fibers were scanned at a step size of 0.05° with 2θ between 5° and 60° . Cellulose samples are routinely analyzed by X-ray diffraction to determine the crystallinity (French, 2014; French and Santiago Cintrón, 2013). The Peak deconvolution was

85 performed by curve fitting to the observed pattern (French, 2020). In particular, subtracting
 86 instrumental background before peak deconvolution ensures a stable baseline fitting and reduces
 87 fitting error (Yao et al., 2020). The crystallinity (x) was calculated using the following equation:

$$88 \quad x = \frac{\sum I_c}{\sum I_c + \sum I_a} \times 100\% \quad (1)$$

89 Where $\sum I_c$ represents the peak diffraction integral intensity crystalline region of the samples, and
 90 $\sum I_a$ represents the intensity scattered by the amorphous fraction of the material.

91 The surface energies of samples, including surface tension, and polar and dispersion-component
 92 were calculated based on Owens–Wendt–Rabel–Kaelble method (OWRK). The slope and intercept
 93 of fitting line with three different liquids (water, ethylene glycol, ethanol) were obtained using the
 94 OWRK method (Table 1). The OWRK method expresses the surface energy between liquid and solid
 95 using the following equations:

$$96 \quad \delta_l(1 + \cos \theta) = 2 \left(\sqrt{\delta_s^d \delta_l^d} + \sqrt{\delta_s^p \delta_l^p} \right) \quad (2)$$

$$97 \quad \frac{(1 + \cos \theta)}{2} \cdot \frac{\delta_l}{\sqrt{\delta_l^d}} = \sqrt{\delta_s^p} \cdot \sqrt{\frac{\delta_l^p}{\delta_l^d}} +$$

$$98 \quad \sqrt{\delta_s^d} \quad (3)$$

99 where δ_l is surface energy of liquid, δ_s is surface energy of solid, $\cos \theta$ is the contact angle, while
 100 superscript p and d represent polar and dispersion components, respectively. (Kaelble, 1970; Murray
 101 et al., 2012; Owens and Wendt, 1969; Rabel, 1971). Eq. (3) can be further expressed into slope and
 102 intercept as follows:

$$103 \quad y = \sqrt{\delta_s^p} \cdot x + \sqrt{\delta_s^d} \quad (4)$$

$$104 \quad x = \sqrt{\frac{\delta_l^p}{\delta_l^d}} \quad (5)$$

105 **Table 1** Properties of liquids for surface energy measurement of fibers.

Liquid type	Surface tension (mN/m)	Polar component ^a (mN/m)	Dispersion component ^a (mN/m)
Water	72.1	52.2	19.9
Ethylene glycol	48.0	29.0	19.0
Ethanol	22.1	4.6	17.5

106 ^a Using figure from liquids database of tendionmeter (DCAT11, Dataphysics, Germany)

107 Chemical composition

108 Composition analysis of samples were performed according to standardized methods of fibrous
109 materials. The proportions of the chemical composition were based on GB/T standard methods.
110 Holocellulose (sum of the cellulose and hemicelluloses), acid-soluble lignin and Klason-lignin were
111 determined according to GB/T 2677.10-1995, GB/T 2677.8-94 and GB/T 10337-2008, respectively.
112 The cellulose of samples was quantified using TAPPI standard methods, followed by averaging of
113 the results and comparison among experiments.

114 Characterization of Human-like collagen

115 Two types of human-like collagen, Trauer (Guangzhou Trauer Biotechnology Co., Ltd.) and
116 MeiQ (Yangzhou Merry Bio-Technology Co., Ltd.), was employed in this experiment, with the pure
117 water as blank simple. The densities and surface tensions of three liquids were measured by a tension
118 meter (DCAT11, Dataphysics, Germany), while viscosities were determined using a viscosimeter
119 (DV2TLV, Brookfield, USA) at a temperature of 25°C, with a spindle speed of 200 rpm and a rotor
120 (SC4-18(18)). The pH of liquids was determined by pH-220B from China. The detailed properties of
121 experimental liquids are shown in Table 2.

122 **Table 2** Properties of liquids for experiment

Solution type	Density (g/cm ³)	Viscosity (mPa s)	Surface tension (mN/m)	pH
Trauer	1.00	1.65	48.14	5.42
MeiQ	1.01	20.98	37.90	5.33
Water	1.01	1.07	68.38	7.18

123 Adsorbed morphology

124 The adsorption process on the fiber surface was observed and recorded using a microscope
125 consisting of an optical contact angle meter OCA15EC. First, the fiber was placed on the sample
126 holder, and the horizontal screw rod was regulated to ensure that the fiber was in a horizontal state.
127 Second, 1 to 10-micron droplets were sprayed using the humidifier, which was directed onto the fiber

128 from a tube (3cm in diameter) to form droplets followed by the condensation of small droplets into
129 larger droplets. Finally, the adsorbed morphology was observed and recorded with the optical
130 microscope and CCD components.

131 The fiber hollow cavity was used for droplet penetration by taking a few samples on a glass slide
132 and dropping the droplets using a disposable pipette. The penetration of droplets into the fiber hollow
133 cavity was captured by a polarizing microscope (ECLIPSE LV 100 POL, Nikon, Japan).

134 Evaluation of liquid sorption rate

135 As shown in Fig. 1, the adsorption experiment was carried out using the tensiometer DCAT11.
136 First, the sample was evenly distributed into a cylindrical container (2) that consisted of a length of
137 40mm and an inner diameter of 12mm. The sample filling mass, length and density were 0.2 g, 20
138 mm and 0.084 g/cm^3 , respectively. Then, the sample container (2) was hung under the microbalance
139 using a unique sample rack. The beaker containing the test liquid was placed in the constant
140 temperature liquid reservoir (3) on the elevator (5). The temperature of the test liquid was set to 25°C
141 through a thermostatic bath. Finally, recording was done by opening computer (6) and testing the
142 software performance. The change in absorbed mass was recorded using the microbalance (1) at the
143 same intervals and plotted in the form of a curve using the computer (6).

145 **Fig. 1.** Diagram of device for adsorption

146 Results and discussions

147 Characterization of fiber

148 Morphology and structure

149 The morphologies of Kapok fibers (raw and different alkali modification) are illustrated in Fig.
150 2. Because of the plant wax coverage, the raw Kapok fiber shows a cylindrical shape with a smooth
151 surface (Fig. 2(a)). Following alkali modification (Fig. 2(f-j)), the fiber surface becomes rough with
152 vertically arranged grooves due to the shedding of surface plant wax. As the modified alkali
153 concentration increases from 2% to 8%, severe damage on the surface structure was observed on
154 AKF-8(Fig. 2(j)), consisting of apparent traces of alkali corrosion.

155 **Fig. 2.** SEM image of surface morphologies in Kapok fiber(a), AKF-2(c)(d), AKF-4(f), AKF-6(g) and AKF-8(h)
156 and the cross-section in Kapok fiber(b) (g) and AKF-2(d).
157

158 Fig. 2(b) (d) shows the cross-sections of raw Kapok fiber and AKF-2. The raw Kapok fiber has
159 a hollow cavity structure consisting of numerous multilevel buckling micropores in the cell wall Fig.
160 2(e). Several studies have found excellent oil absorption capacity in the Kapok fiber largely because
161 oil can easily enter the hollow cavity through the open ends of the fiber wall and through the multi-

level buckling micropores on cells (Dong et al., 2015a) (Dong et al., 2014). As shown in Fig. 2(d), the AKF-2 still depicts the hollow cavity structure with a thinner cell wall, indicating that the fibril structure is still intact. When adsorption occurs, the liquid can still enter the hollow cavity through the open ends of the fiber wall. Previous studies (Wang et al., 2012) have also shown that the appropriate NaOH modification is beneficial for improving absorbency depending on the modification concentration, temperature and time.

Composition analysis

The absorption peaks at $3400 \sim 3200 \text{ cm}^{-1}$ and $2918 \sim 2901 \text{ cm}^{-1}$ are attributed to OH and C-H's stretching vibration, respectively. These peaks are characteristics of the presence of cellulose (Andreeva et al., 2002). Fig.3(a) shows that the raw Kapok fiber has two absorption peaks at 3335.36 cm^{-1} and 2919.35 cm^{-1} . However, no significant changes were observed as the alkali concentration increased, indicating no damage to the cellulose of the Kapok fiber. The absorption peaks at 1736.14 cm^{-1} , 1593.44 cm^{-1} and 1239.33 cm^{-1} are attributed to the carbonyl group, the aromatic ring skeleton group, and the phenolic hydroxyl group; respectively, which are characteristics of absorption peak for lignin. It can be seen from Fig.3(a) that the absorption peaks start to disappear at 1736.14 cm^{-1} and 1239.33 cm^{-1} , and the absorption peak became weaker at 1593.44 cm^{-1} , indicating the partial removal of lignin. The absorption peaks about alkanes (1371.00 cm^{-1} and 2919.35 cm^{-1}) become weaker after alkali modification.

Fig.S1 shows that the diffraction peaks of raw fiber and alkali-modified Kapok fiber have no significant fluctuations, around 15.8° , 22.4° and 34.6° 2θ . It indicates that the crystal structure of cellulose has not changed by alkali-modified, and the profile has been considered as the pure amorphous cellulose XRD profile (Yao et al., 2020). And the Fig.S2 shows the peak deconvolutions of raw fiber and alkali-modified Kapok fiber, the crystallinity using the ratio of the crystalline peak areas and the total XRD profile area were calculated by peak deconvolution method after subtracting the background intensity. Fig.3(b) shows that the raw fiber's crystallinity is 35.71%, which is likely due to presence of amorphous components including lignin, hemicellulose, and wax. The crystallinity increased from 35.71% to 48.22% following the increase in alkali concentration from 2% to 6%. Further increases in alkali concentration from 6% to 8% resulted in dropping fiber's crystallinity from

190 48.22% to 43.94%. The increase in crystallinity after alkali treatment can be associated with the
 191 dissolution of free molecular chains in the amorphous region. The dissolution of free molecular chains
 192 increased the proportion of crystalline regions, thereby increasing the crystallinity.

Fig. 3. FTIR spectra (a), Crystallinity (b) and Chemical Component(d) of Kapok, AKF-2, AKF-4, AKF-6 and AKF-8 and Yield(c) of AKF-2, AKF-4, AKF-6 and AKF-8.

196 The results obtained by examining the yield and chemical components of alkali-modified Kapok
 197 fiber are summarized in Fig. 3(c)(d). As shown in Fig. 3(c), the yield decreases from 71.2% to 59.5%
 198 as the alkali concentration increases from 2% to 8%. Likewise, the raw Kapok fiber mainly consists
 199 of cellulose (39.95%), hemicellulose (37.01%), acid-soluble lignin (11.09%) and Klason lignin
 200 (4.46%) (Fig. 3d). Results also show that the chemical composition of different alkali-modified
 201 Kapok fibers has a substantial effect on the hemicellulose and lignin content. For example, 38.17%
 202 of hemicellulose and 39.46% of acid-soluble lignin were removed in AKF-2, while 69.46% of

203 hemicellulose and 73.54% of acid-soluble lignin were removed in AKF-8. However, our results show
 204 that the proportion of cellulose and Klason lignin experience an upward trend. The results indicate
 205 that the alkali dissolves part of the hemicellulose and lignin; however, the cellulose and Klason lignin
 206 are insensitive to the alkali concentration.

207 **Surficial wettability**

208 **Fig. 4.** Contact angle of Kapok, AKF-2, AKF-4, AKF-6 and AKF-8 on Water (a), MeiQ (b) and Trauer (c),
 209 respectively. Surface energy (d) of Kapok, AKF-2, AKF-4, AKF-6 and AKF-8.
 210

211 To characterize the surface property of fibers, the contact angle of liquid was measured. As
 212 shown in Fig. 4(a), the raw Kapok fiber showed the water contact angle was 129.2°, which is
 213 considered as a hydrophobic surface. However, the water contact angle decreased below 50° with
 214 decreasing concentration of alkali (2 wt%: 48.3°, 4 wt%: 44.6°, 6 wt%: 45.2°, 8 wt%: 44°), the AKF-
 215 2, AKF-4, AKF-6, and AKF-8, which is considered as hydrophilic. The contact angle of human-like

collagen showed a similar trend (Fig. 4(b)(c)). The lower contact angle achieved in alkali-modified fiber was supposed to be related with the surface energy. As depicted in Table S1, the surface energy of fiber rose rapidly from 7.80mN/m to 125.05-140.06mN/m with polar component playing a central role in stimulating the surface energy. On the other hand, Fig. 4 (d) shows that the surface energies increase with an increase in NaOH concentration from 2% to 4%, with the highest surface energy of 140.06mN/m at 4% NaOH concentration. However, there was a downward trend in surface energies with an increase in NaOH concentration from 4% to 8%. Generally, the higher surface energy of the substance indicates the better wettability of water on the surface and the smaller contact angle of water. Ko et al. (Ko et al., 1981) also suggested that the surface energy increases with the rise in hydrophilic compared to the hydrophobic groups. In this study, the NaOH-modification removed the surface wax and exposed hydrophilic hydroxyl groups, which played an important role in hydrophilicity and fiber-matrix adhesion.

Characterization of adsorption capacity

Fig. 5(a)-(b) shows the equilibrium adsorption amount (q_e) and equilibrium time (t_s) of fibers. The absorption curves of water and human-like collagen to alkali-modified Kapok fibers are shown in Fig. 5(c)-(f). Results showed that the adsorption amount of raw Kapok fiber is around 0.6g/g, which accounts for only one twenty-first of the adsorption amount of alkali-treated Kapok. Given that the adsorption amount was almost negligible, we did not proceed with further analysis of the adsorption curve. The equilibrium time of AKF-2 to MeiQ was 1399.80s, while that of Water and Trauer were only 274.80s and 300.00s, indicating that Water and Trauer had an adsorption time that was shorter than MeiQ by approximately 4-5 times. The equilibrium adsorption amounts of water, MeiQ and Trauer were 12.61g/g, 11.96 g/g and 12.10 g/g; respectively, without significant variation. Similar trends were found in the AKF-4, AKF-6 and AKF-8.

It is likely that the difference in equilibrium time is related to changes in liquid viscosity. As illustrated in Table 1, water and Trauer have similar liquid viscosity resulting in similar adsorption times. However, the viscosity of MeiQ's (20.98/mPa) was 20 times higher than water and Trauer. The fiber has small particle size and weak adhesion to low-viscosity liquids, but large particle size and strong adhesion to high-viscosity liquids (Dong et al., 2015a). MeiQ with high viscosity is more likely

244 to be around the Kapok assembly's pores, which can prevent the liquid from entering the interior.

Fig. 5. Adsorption curves of AKF-2, AKF-4, AKF-6 and AKF-8 on Water (a), MeiQ (b) and Trauer (c), respectively, Symbols represent an average of three trials and error bars plotted as a standard deviation.

247 However, it is noteworthy that the equilibrium adsorption time had inverse relationship with the
 248 alkali concentration. The equilibrium time of AKF-4 for MeiQ was 169.80 s, which was shorter than
 249 that of AKF-2 by 49.97%. The adsorption time in AKF-6 showed similar trend as AKF-4, with AKF-
 250 6 shorter than AKF-2 by 42.99%. For AKF-8, there were more significant gaps in equilibrium
 251 adsorption time, but the gaps were shortened at 24.98% for the MeiQ. The adsorption time showed
 252 similar tendency for water and Trauer, which was likely due to an increase in the alkali concentration
 253 that exposed more hydrophilic groups due to the removal of fiber surface wax. Compare to the
 254 equilibrium time, the equilibrium adsorption amount of AKF-2 on three different liquids was similar
 255 under the same filler density (0.0884 g/cm³), with only an 0.65 g/g discrepancy between the smallest
 256 in MeiQ and the largest in water. A similar result appears in AKF-2, AKF-4, AKF-6 and AKF-8 to the
 257 adsorption of liquids. Besides, as illustrated in Table S2, different alkali-modification seems to exert
 258 negligible influence on the equilibrium adsorption amount for three liquids, with only 0.1g/g
 259 difference for AKF-2 and AKF-8. Several studies have reported that the oil absorbency of the Kapok

assembly's depend on effective inter-fiber pore volumes and hollow cavity structure (Dong et al., 2015a) (Abdullah et al., 2010) (Rengasamy et al., 2011). Therefore, it is likely that the four fibers mentioned above have a minimal difference in equilibrium adsorption amount largely due to same filler density (0.0884 g/cm³) and pore volume.

Absorption kinetics

To further study the human-like collagen adsorption mechanism of AKF, the Kapok fibers' cumulative adsorption data treated under different alkali concentrations were fitted using the mathematical model of adsorption kinetics. The adsorption kinetic process of solid to liquid phase is generally described using the Quasi-first-order and Quasi-second-order kinetic model (ZengxiaoCai et al.) (Tanzifi et al., 2018), and Weber-Morris internal diffusion model (Hubbe et al., 2019) (Haque et al., 2020) (Bortoluz et al., 2020).

1) Quasi-first-order kinetic model

The Quasi-first-order kinetic model is a first-order rate equation based on solid adsorption, which is applied to liquid phase adsorption. It is one of the most common models which can be expressed using the following equation:

$$\log(q_e - q_t) = \log q_e - \frac{k_f}{2.303} t \quad (6)$$

Where q_e (g/g) is the equilibrium adsorption amount, q_t (g/g) is the adsorption amount at time t and k_f is the first-order adsorption rate constant.

2) Quasi-second-order kinetic model

It is based on the assumption that the chemical adsorption mechanism controls the adsorption rate. The chemical adsorption mainly refers to the electron sharing and electron transfer between the adsorbent and adsorbate. The following equation is used to express the Quasi-second order kinetic model:

$$\frac{t}{q_t} = \frac{1}{k_s q_e^2} + \frac{1}{q_e} t \quad (7)$$

Where k_s represents the second-order adsorption rate constant.

3) Weber-Morris model

The Weber-Morris internal diffusion model is often used to describe the process of adsorbate

diffusion in the adsorbent pores. The model first analyzes the control steps in the reaction, followed by calculation of the diffusion rate constant in the adsorbent particles. The following equation is used to calculate the diffusion:

$$q_t = k_{ip}t^{1/2} + C \quad (8)$$

Where C is a constant related to the thickness and boundary layer, k_{ip} is internal diffusion adsorption rate constant.

The Quasi-first-order and Quasi-second-order kinetic models' Parameters are exhibited in Table 3, where $q_{e,exp}$ and $q_{e,cal}$ represents the equilibrium adsorption amount estimated from the experiment and model; respectively, while R represent the fitting coefficient. Results show that the Quasi-first-order kinetic model had a better fitting effect than the Quasi-second-order kinetic model for water, primarily due to closer equilibrium adsorption amount ($q_{e,exp}$ and $q_{e,cal}$) and higher R^2 values. However, the fitting effect for human-like collagens indicated that the adsorptions were aligned with both Quasi first- and second-order kinetic models (Fig. 6). Herein, it is more likely that the fiber's adsorption to water is the physisorption mechanism rather than the chemisorption, resulting from Van Der Waals, while the adsorption of human-like collagen is due to the coexistence of physisorption and chemisorption. In the Quasi-first-order kinetic model, the rate constant (k_f) of three liquids decreases with increase in liquid viscosity. Water has the largest rate constant because of its lowest viscosity (1.07mPa s). Compare to lower viscosity liquids, the higher viscosity liquids are more difficult to diffuse under small sized pores of the fiber assembly. The k_s in the Quasi-second-order kinetic model of human-like collagens show a similar trend as k_f . Overall, results showed that the higher viscosity liquid has a stronger interaction force on fiber compared to the lower viscosity liquid.

Table 3 Kinetic parameters for Pseudo-first-order and Pseudo-second-order.

Adsorbate	Adsorbent	$q_{e,exp}(g\ g^{-1})$	Pseudo-first-order			Pseudo-second-order		
			$q_{e,cal}(g\ g^{-1})$	$k_f(s^{-1})$ (10^{-3})	R^2	$q_{e,cal}(g\ g^{-1})$	$k_s\ (10^{-3})$	R^2
AKF-2	Water	12.615	12.364	21.991	0.845	6.854	10.962	0.572
	Trauer	11.965	15.528	18.400	0.995	15.318	0.817	0.941
	MeiQ	12.105	13.564	3.039	0.998	15.413	0.174	0.957
AKF-4	Water	12.630	12.406	36.251	0.992	35.842	0.774	0.223
	Trauer	11.440	9.153	25.056	0.983	13.349	2.523	0.992

	MeiQ	12.063	12.316	7.254	0.992	13.762	0.556	0.989
	Water	12.583	12.365	51.112	0.992	21.353	2.178	0.872
AKF-6	Trauer	11.309	8.574	55.018	0.982	12.766	6.673	0.992
	MeiQ	11.869	12.533	8.198	0.975	12.755	1.316	0.998
	Water	13.055	12.299	114.632	0.984	18.382	2.949	0.887
AKF-8	Trauer	12.014	8.592	36.341	0.984	13.430	4.603	0.992
	MeiQ	12.018	12.552	8.544	0.982	13.518	0.957	0.992

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311

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Fig. 6. Quasi-first-order(a-c) and Quasi- second-order kinetic(d-f) model plots of alkali-modified Kapok fiber.

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319

The Weber-Morris model was further used to evaluate the contribution of surface and intraparticle diffusion in the adsorption process. Suppose that the relationship between q_t and $t^{1/2}$ is not only linear but also passes through the origin. In that case, the Weber-Morris pore diffusion model believes that intraparticle diffusion is assumed to be controlled by a sole rate. As shown in Fig. 7, q_t and $t^{1/2}$ have a linear relationship, but does not pass through the origin, indicating that the intraparticle diffusion played a positive role in the adsorption steps. However, intraparticle diffusion is not the only step to control the adsorption process because there is other simultaneous adsorption mechanism

that control adsorption. The curves of q_t and $t^{1/2}$ are divided into three linear stages (Fig. 7), which implies that there are three consecutive adsorption steps. In the three consecutive adsorption stages of AKF-2, the adsorption capacity of the liquid accounted for 7.42-19.23%, 64.38-84.61% and 5.55-16.39%; respectively, of the total adsorption. Among the three adsorption stages, the second is the main adsorption stage, which resulted in the adsorption of about 57.21-81.08%, 43.78 -70.57% and 55.32-75.92% by AKF-4, AKF-6 and AKF-8, respectively. The K_{ip} value decreases with an increase in human-like collagen's viscosity (Table 4), indicating that lower viscosity can promote diffusion in the fibers. Herein, the adsorption kinetics of AKF to human-like collagen is attributed to both surface and intra-particle diffusion, with the latter playing a more dominant role as the main form of diffusion.

Fig. 7. Weber-Morris model plots of AKF-2, AKF-4, AKF-6 and AKF-8 on Water (a), MeiQ (b) and Trauer (c), respectively.

Table 4 Parameters for Weber–Morris models.

Adsorbate	Adsorbent	Weber-Morris								
		First stage			Second stage			Third stage		
		C_1	$k_{ip1}(\text{g g}^{-1} \text{s}^{-1})$	R_l^2	C_2	$k_{ip2}(\text{g g}^{-1} \text{s}^{-1})$	R_2^2	C_3	$k_{ip3}(\text{g g}^{-1} \text{s}^{-1})$	R_3^2
AKF-2	Water	0.468	0.199	0.850	16.079	2.030	0.994	8.113	0.284	0.933
	Trauer	0.023	0.267	0.948	3.014	1.210	0.985	8.848	0.182	0.886
	MeiQ	0.178	0.161	0.963	2.473	0.512	0.987	7.101	0.133	0.925
AKF-4	Water	0.165	0.183	0.963	7.980	2.328	0.977	9.891	0.214	0.902
	Trauer	0.133	0.698	0.986	0.989	1.361	0.978	9.973	0.099	0.942
	MeiQ	0.072	0.382	0.988	0.247	0.584	0.952	7.391	0.165	0.923
AKF-6	Water	1.039	1.135	0.924	4.775	2.237	0.990	9.512	0.290	0.828
	Trauer	0.143	0.965	0.995	1.103	2.092	0.970	10.308	0.093	0.820
	MeiQ	0.219	0.720	0.972	2.846	0.464	0.962	10.034	0.064	0.777
AKF-8	Water	0.728	1.384	0.959	5.815	3.886	0.968	11.835	0.168	0.845

Trauer	0.079	0.668	0.978	1.294	1.880	0.947	11.024	0.076	0.920
MeiQ	0.038	0.553	0.9983	0.4136	0.660	0.957	8.444	0.146	0.931

333 Adsorption mechanism

334 As shown in Fig. 9, the adsorption could be broadly divided into three stages which includes
335 surface-, inter-fiber-, and hollow cavity-diffusion. In the first step (i.e., surface diffusion) alkali
336 modification can stimulate the removal of surface wax and expose the hydrophilic hydroxyl groups,
337 which is advantageous in fiber-matrix adhesion and wettability. Compared to raw Kapok fiber, the
338 human-like collagen diffuses on the AKF-2's surface and slowly forms larger liquid droplets (Fig.
339 8(a)(b)). In the second step (i.e., inter-fiber diffusion) a wicking phenomenon occurred under the
340 capillary action of the inter fiber gap to liquid, resulting in the diffusion of adsorbed molecules diffuse
341 to the depths. In the third step (i.e., fiber hollow cavity diffusion), Fig. 8(c)(d) shows that the liquid
342 molecules enter the fiber's hollow cavity because of the presence of numerous multi-level buckling
343 micropores on cell wall and the open ends of the fiber wall, through which the liquid enter the fiber's
344 hollow cavity. These micropores connect the liquid between the hollow cavity and fiber's surface,
345 forming a microporous channel for liquid flow. It is important to note that the second and third step
346 of the adsorption process is intra-particle diffusion caused by the capillary action of effective pore
347 volume and hollow cavity on the liquid. The Kapok fiber's absorption shows a dual-scale liquid
348 adsorption behavior that occurs through both inter-fiber pores and fiber's hollow cavity. The dual
349 adsorption behavior is the primary reason for better adsorption capacity of the Kapok fiber. The force
350 of adsorption is mainly manifested as an orientation force because of the polar macromolecule
351 characteristics of Kapok fiber, and the presence of great dipole moment in the human-like collagen.
352 When two polar molecules approach each other, the molecules will rotate and arrange in a specific
353 direction due to the repulsion of the same polarity, followed by electrostatic attraction. Therefore, the
354 orientation force plays an important role in generating the electrostatic attraction between the
355 permanent dipole of polar molecules.

Fig. 8. Adsorption morphology of human-like collagen on the fiber surface, raw Kapok(a) and AKF-2(b). Micrograph of AKF-2 in nature state (a), water (b) penetrating fiber' hollow cavity.

Fig. 9. The three processes of the adsorption of liquid, (a) surface diffusion, (b) inter-fiber diffusion and (c) fiber' hollow cavity diffusion.

The FTIR spectra of AKF-2 and human-like collagen were investigated before and after adsorption to further explore the existence of adsorption mechanisms. The collagen is a polypeptide chain composed of α -amino acids formed through peptide bonds. The α -carboxylic and α -amino at

365 both ends of the polypeptide chain can receive or release protons (Han et al., 2019; Pawelec et al.,
 366 2016). In the process of adsorption, these groups can form ionic and covalent bonds with hydroxyl
 367 and carboxyl groups on the fiber. And the collagen contains a large number of carboxyl, amino and
 368 hydroxyl groups, cellulose contains hydroxyl and a small amount of carboxyl groups. The carboxyl,
 369 amino and hydroxyl in collagen and the non-ionic area's hydroxyl groups on the plant fiber's surface
 370 can form numerous intermolecular hydrogen bonds (Lohrasbi et al., 2020). As show in Fig. 10(a),
 371 three absorption peaks at 923.57 cm^{-1} , 859.80 cm^{-1} and 993.63 cm^{-1} , corresponding to the C-H out-
 372 of-plane bending vibration of MeiQ, and 2880.88 cm^{-1} , corresponding to C-H stretching vibration,
 373 diminish and disappear after adsorption. The absorption peak at 3304.51 cm^{-1} , corresponding to the
 374 O-H stretching vibration of AKF-2, slightly shifted, intensified and expanded after adsorption. In
 375 comparison (Fig. 10(b)), The absorption peak at 3304.51 cm^{-1} , corresponding to the O-H stretching
 376 vibration of Traure, slightly shifted and intensified after adsorption. Two absorption peaks at 1554.20 cm^{-1} ,
 377 cm^{-1} , corresponding to the C=C stretching of Traure, and 1039.80 cm^{-1} , corresponding to the C-C
 378 stretching of Traure, amplify after adsorption, while the absorption peak at 1657.96 cm^{-1} ,
 379 corresponding to the C=C stretching of AKF-2, diminish after adsorption. Herein, results show that
 380 the physisorption process was primarily accompanied by chemisorption.

381
 382 **Fig. 10.** FTIR spectra of AKF-2 before and after MeiQ adsorption(a) and Trauer adsorption (b)

383 Conclusion

384 We investigated the adsorption kinetic, equilibrium, and mechanistic characteristics of human-
 385 like collagens using different alkali-modified Kapok fibers. Based on our findings, we provide five

major conclusions that would likely improve our understanding of kinetic, equilibrium and mechanistic features of human-like collagens. (1) The characterization of the AKF showed that the fiber's surface morphology was strongly dependent on alkali concentration, where 2% of modified NaOH concentration did not have a significant effect on the hollow cavity structure. The crystallinity increased from 35.71% to 48.22% following the increase in alkali concentration from 2% to 6%. Further increases in alkali concentration from 6% to 8% resulted in dropping fiber's crystallinity from 48.22% to 43.94%. The raw Kapok fiber mainly consists of cellulose (39.95%), hemicellulose (37.01%), acid-soluble lignin (11.09%) and Klason lignin (4.46%), and the part of hemicellulose and lignin were removed through alkali modification. (2) With further increase in alkali concentration, there is a rapid loss of Kapok fiber's surface energy increase from 7.80 mN/m to 125.05-140.06. mN/m, while the water contact angle decreased from 129.2° to 48.3-44°. The contact angles of human-like collagens showed a similar trend. (3) The adsorption amount of raw Kapok fiber is around 0.6g/g, which accounts for only one twenty-first of adsorption amount of the alkali-treated Kapok. As the NaOH concentration increase, there was a decrease in adsorption time of human-like collagen time. In contrast, the adsorption time increased with high viscosity. This is primarily due to strong adhesion and large adhesion particle sizes that inhibited diffusion. However, the adsorption amount is not sensitive to alkali concentration because of the same filler density (0.0884 g/cm³) and pore volume in the Kapok assembly. (4) The absorption data of human-like collagen were described well with both Quasi-first-order and Quasi-second-order kinetic models, indicating the existence of both physisorption and chemisorption. The data also exhibited a better fit to the Weber-Morris model, indicating that the intra-particle diffusion played a positive role in the adsorption steps. (5) The adsorption step could be broadly divided into three stages which include surface, inter-fiber, and hollow cavity-diffusion.

Compliance with ethical standards

Conflicts of interest: The authors declare that they have no conflict of interest.

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