

Polyurethane foams synthesis with addition of banana peel and application as adsorbent of tartrazine dye

Síntese de espumas de poliuretano com adição de casca de banana e aplicação como adsorvente do corante tartrazina

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Abstract

The purpose of this work was to synthesize, characterize and evaluate the adsorption capacity of polyurethane foams prepared with addition of banana peel waste (BPW) as a sustainable substituent for the polyol. Studies were carried out with pure polyurethane (PU) and the composite with 10% (w/w) of the waste (PUB) and its adsorption capacities were evaluated with tartrazine yellow dye. The thermogravimetric analysis showed the thermal stability of both the waste and its composite. Fourier transform infrared spectroscopy indicated similar bands in the foams. It was observed by scanning electron microscopy that foams without waste consists of compacted canals, whereas foams with banana peel are formed by spheroid regions. The adsorption experiments were performed both in acidic and basic media and the best adsorption results were obtained at pH 3.0, with a percentage of adsorption of 99.7% for PU and 87.3% for PUB. The kinetic study showed that the highest values of adsorption were reached in 120 min for both types of foam and the process was better adjusted within the pseudo-second-order kinetic model. From this study, the biocomposite of polyurethane with banana peel could be obtained applying renewable material, low costs, high efficiency in adsorption of an industrial dye and potential use for treatment of effluents.

Keywords: adsorption, banana peel, waste, polyurethane, composite.

Resumo

O objetivo deste trabalho foi sintetizar, caracterizar e avaliar a capacidade de adsorção de espumas de poliuretano preparadas com adição de resíduos de casca de banana (BPW) como um substituinte sustentável do poliálcool. Os estudos foram realizados com poliuretano puro (PU) e o compósito com 10% do resíduo (PUB) e suas capacidades de adsorção foram avaliadas com corante amarelo de tartrazina. A análise termogravimétrica mostrou a estabilidade térmica do resíduo e de seu compósito. A espectroscopia no infravermelho por transformada de Fourier indicou bandas semelhantes nas espumas. Observou-se por microscopia eletrônica de varredura que as espumas desprovidas de resíduos consistem em canais compactados, enquanto as espumas com casca de banana são formadas por regiões esferoides. Os experimentos de adsorção foram realizados em meios ácido e básico, e os melhores resultados de adsorção foram obtidos em pH 3,0, com percentual de adsorção de 99,7% para PU e 87,3% para PUB. O estudo cinético mostrou que os maiores valores de adsorção foram alcançados em 120 min para ambos os tipos de espuma e o processo foi melhor ajustado dentro do modelo cinético de pseudo-segunda ordem. A partir deste estudo, foi possível obter o biocompósito de poliuretano com casca de banana, empregando material renovável, de baixo custo, com alta eficiência na adsorção de um corante industrial e potencial uso no tratamento de efluentes.

Palavras-chave: adsorção; casca de banana; resíduos; poliuretano; compósito.

INTRODUCTION

Banana is a typical fruit in tropical countries and represents about 40% of the world trade in fruits. Brazil is the sixth most important banana producer (IBGE, 2017). For example, Brazil produced a total of 6.675.100 t of bananas in 2017 (FAOSTAT, 2014), specially *Musa cavendishi*, which is quite used in the food industry although are eaten raw, used in baking, candies, fruit compotes, fruit salads, and to complement foods.

However, some important parts of bananas such as the peels are still often discarded due to lack of knowledge on its usefulness and benefits. The banana peel is the principal fruit residue in the banana food industry and it accounts for 30-40% of the total fruit weight (KUMAR et al., 2012). The banana waste is mainly composed of fiber components such as cellulose (50-60%), hemicelluloses (25-30%), pectin (3-5%) and lignin (12-18%) that allows its application in the development of new products at low cost (KUMAR et al., 2012; IBRAHIMET al., 2010; KORONIS et al., 2014).

The waste that derives from the industrial process of banana, such as leaves, roots and peels can be used for bioenergy production and composting, as well as for effluents treatment, sorbents of dyes and of carcinogenic aromatic composites (GUPTA AND GUPTA, 2015; PAPPU et al., 2015). Studies investigated the use of banana peel waste as sorbent, the removal of strontium (II) from aqueous solution reached the maximum biosorption capacity of 41.5 mg/g at pH 7 in 10 minutes (MAHINDRAKAR AND RATHOD, 2018). Banana, cucumber and potato peels were used as biosorbents for the removal of methylene blue and Orange G, for methylene blue the maximum adsorption capacity was found using banana peel, 211.9 mg/g at pH 6, and for Orange G, banana peel reached 20.9 mg/g of adsorption at pH 2 (STAVRINOUE et al., 2018).

In the latest years, the number of initiatives related to the sustainable destination of agroindustrial

wastes has increased around the world, due to its high availability, biodegradability and low cost (COLPINI et al., 2014; OLIVEIRA et al., 2019; KUMAR et al., 2014; SANTOS et al., 2010, WANG et al., 2014). However, depending on the waste, the adsorption efficiency may not be high due to the decomposition of these natural constituents in aqueous medium, for example by thermic, dissolution and oxidation processes, among others. In this case, an interesting approach is to incorporate these agroindustrial residues into stable polymeric matrices, forming stable biocomposites (ANNADURAI, 2002; MOHAMMADI et al., 2014; ZIA et al., 2015).

Once the synthesis of polyurethane foams and related manufactured products involves high-cost synthetic polyols, such as polyethylene-glycol, one alternative is to incorporate agroindustrial wastes in the foams during the polymeric synthesis. This is possible since banana peel waste has several hydroxyl groups of its constituent carbohydrates that can act as a polyol substitute, used in the industrial synthesis of PU foams. Replacing synthetic polyols by adding biopolymers in the synthesis of polyurethane foams often results in improvement of its physical and chemical properties, reduction of environmental impact in the processing, increase in biodegradability and can lead to the development of new potential biosorbents (AUNG et al., 2014; KARA et al., 2014; OBRUCA et al., 2011; Zia et al., 2015).

In food and drug industrial wastewaters, the Yellow tartrazine dye (TAR), also known as Acid Yellow 23 and E102 additive, is very commonly. It belongs to the group of azo-dyes, which comprises 60% of all dyes used in the world (DAWODU AND AKPOMIE, 2016). As many azo-dyes and products derived from its degradations are toxic and carcinogenic, industrial effluents that contain such type of dye must be properly treated before being discarded to the environment (MATHIEU-DENONCOURT et al., 2014).

In this work, the purpose was to produce polyurethane foams by using banana peel to replace synthetic polyol, and to apply the foams in TAR

adsorption. It was detailed the physicochemical characterization of the foam and also the efficiency of adsorption and kinetics of adsorption.

MATERIALS AND METHODS

Materials

The reagents used were 4,4'-diphenylmethane diisocyanate (MDI) (Sigma-Aldrich), stannous octoate (Sigma-Aldrich), polyethylene-glycol 1500 g mol⁻¹ (PEG 1500) (Synth), glycerol (Synth), polymethylhydrosiloxane (PMHS) and tartrazine dye (Synth). All reagents were analytical grade.

Waste treatment

The banana peel waste was obtained in the local market of Umuarama, a city in the state of Paraná, Brazil. The peel was triturated by an industrial grinder (Becker, Brazil) and dried in an oven (Marconi, Brazil) at 60 °C for two hours. The dried material was shifted with a 48-mesh stainless steel sieve and again dried in the oven, which resulted in the banana peel flour used in the synthesis of polyurethane foams that would partially replace synthetic polyol.

Foams synthesis

For the synthesis of the foam that did not contain banana waste (PU), 6.0 g of PEG 1500 were weighed up in a beaker and molten at 50 °C. After cooling, it was added 0.6 mL of PMHS and 0.10 mL of glycerol. With another beaker, 3.0 g of MDI was molten at 50°C. After it was cooled, 0.30 mL of tin octoate catalyst were added. This mixture joined the content of the first beaker, and it was vigorously agitated until the foam grew. Then, it was left at rest for 24 h at room temperature. This same procedure was carried out to the synthesis of the foam containing banana peel (PUB), but using different amounts of banana waste flour to substitute PEG 1500 (5-20%).

Foams characterization

Thermogravimetric analysis (TGA) was

performed by using a Netzsch-Gerätebau GmbH - STA 409 PC Luxx Simultaneous thermal analyzer from room temperature to 800°C, under nitrogen flow of 30 mL min⁻¹. The weight of each sample was approximately 15 mg. ATR-FTIR spectra were obtained with a spectrophotometer Bruker Alpha with diamond crystal of individual reflection, from 400 to 4500 cm⁻¹ and resolution of 1 cm⁻¹. Spectroscopic analysis in the UV-Vis region was performed in the range of 200 to 700 nm with a Thermo Scientific Evolution device 60 using 1 cm light path quartz cuvettes. Scanning electron microscopy (SEM) micrographs were carried out using a scanning electron microscope FEI Quanta 250 at an accelerating voltage of 20 kV. Samples were sputter coated with gold to allow cross-section visualization.

In order to determine the zero point of charge (pH_{ZPC}), it was mixed 0.3 g of foam with 50 mL of KCl 0.01 mol/L solution, prepared with deionized water recently boiled, in eleven erlenmeyers. We adjusted the pH values of each solution to 2.0, 3.0, 4.0, 5.0, 6.0, 7.0, 8.0, 9.0, 10.0, 11.0 e 12.0, by measuring the initial pH of each solution and after agitating it all in a shaker at 120 rpm for 24 h. It was measured the pH_{ZPC} from the graph of final versus initial pH values, when the curve results in zero of difference in those two values.

Tar adsorption by foams produced

To evaluate the pH influence in TAR adsorption, 0.3 g of the foam produced (PU or PUB) was added to 20.0 mL of aqueous solutions of yellow tartrazine (35.0 mg L⁻¹) with pH between 3.0 and 8.0. The samples were kept under agitation in a shaker (Marconi, Brazil) at 120 rpm for 8 h. Then, they were filtered and it was determined the absorbance at 428 nm in spectrophotometer (Femto, Brazil). The amount of dye adsorbed (Q) for each pH was determined according to Eq. 1, being C_i the initial concentration and C_f the final concentration of the TAR solution (mg L⁻¹); m_{foam} is the foam mass used in the adsorption (g).

$$Q = \frac{(C_i - C_f) \cdot 0,02}{m_{foam}} \quad (1)$$

In the kinetic study, 0.3 g of foam (PU and PUB) was added to Falcon flasks containing 20.0 mL of the 35.0 mgL⁻¹TAR solution at pH 3.0 and 6.0. The samples were agitated in a shaker at 120 rpm for 15, 30, 60 and 120 min, and then filtered and analyzed in a spectrophotometer at 428 nm. From the amount of TAR adsorbed by the foams at each time period, obtained by Eq. 1, it was used the pseudo-first-order (Lagergren equation) and pseudo-second-order models, Eq. 2 and Eq. 3, respectively, in order to analyze adsorption kinetic behavior. Such models are used to describe the adsorption of dyes, pigments and other pollutants in solid sorbents, aiming to understand the adsorption mechanism in synthesized composites (STUMM, 1990; GURSES et al., 2006).

$$\ln Q_e = \ln(Q_e - Q_t) + k_1 t \quad (2)$$

Where Q_t represents the biosorption capacity at time t , (mg g⁻¹). The plot of $\ln(Q_e - Q_t)$ against t should present a straight line to confirm the applicability of the kinetic model. k_1 is a constant of pseudo-first-order biosorption (min⁻¹), Q_e represents the biosorption capacity at equilibrium (mg g⁻¹), obtained by adjusting the model, which were determined from the slope and intercept of the plot, respectively.

$$t/Q_t = 1/k_2 Q_e^2 + t/Q_e \quad (3)$$

Where k_2 is a constant of pseudo-second-order biosorption (g mg⁻¹ min⁻¹). Calculated the values of k_2 and Q_e from the plot of t/Q_t against t , and should give a linear correlation (R^2) for the applicability of the pseudo-second-order kinetic model.

RESULTS AND DISCUSSIONS

Characterization of the foams

Different amounts of banana waste flour were tested to replace polyol (PEG 1500), which is commonly used in the foam synthesis. The addition of amounts up 10% (w/w) of BPW in substitution of PEG

1500 did not result in good foams, since flour distribution was not homogeneous and foam size was not as big as it was at 10%. Thus, it was used 10% of BPW in order to replace polyol in the synthesis of the biocomposite.

The interactions of BPW with a polymeric matrix is strongly influenced by its chemical composition. This waste has fibers as main components (80-90%) and also present wide chemical variety of fibers composition (cellulose, hemicellulose, lignin), that can affect the thermal behavior of the biocomposite (Moigne et al., 2011). TGA shows the thermal stability of the waste and its composite (Fig. 1a).

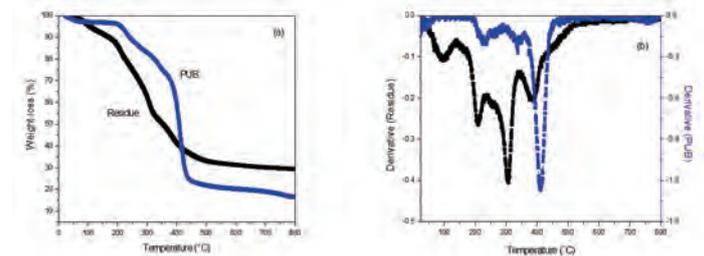


Figure 1 - Thermogravimetric curves of the banana peel waste and the polyurethane foam with banana peel (PUB), under N₂ 30 mL/min flow, a) TG and b) DTG.

In the DTG curves showed by Fig. 1b, it can be seen the degradation of lignocellulosic constituents in PUB, occur with the incidence of four peaks. The first degradation stage of the banana peel waste takes place at 99°C due to the elimination of water from the fibers (BISWAS et al., 2017; CHAIWONG et al., 2013). The second stage occurs at 212°C and its attributed to hemicellulose decomposition; the third one occurs at 309°C, due to cellulose decomposition; as for the last stage, it happens at 386°C and represents lignin decomposition (BEALL AND EICKNER, 1970; CHAIWONG et al., 2013).

PUB presented a discreet peak at 230°C due to the rupture of the weak urethane linkages which dissociated in isocyanates and alcohols, and also of the polyol groups. Main PUB degradation is verified at 413°C because of the products that resulted from the dimerization or trimerization of thermally stable isocyanates that deteriorate at high temperatures,

thus creating volatile products and a small amount of carbonaceous char (BERTA et al., 2006; WU et al., 2018).

The temperature values of maximum mass loss (T_{max}), initial temperature of mass loss (T_{onset}) and residual mass at 800°C (RM800) are presented in Table 1.

Sample	T_{max} (°C)	T_{onset} (°C)	RM800 (%)
Banana peel waste	99	180	29
	212		
	309		
	386		
PUB	230	375	16
	413		
PU (Góes et al., 2016)	380	346	12
	435		

PU - foam without banana peel; PUB - foam containing banana peel.

Rm800 values stand for 29% in the waste and 16% in PUB, which is related to the level of inorganic material in such materials.

In another study carried out with PU it was observed, T_{onset} of 346°C (GÓES et al., 2016). Therefore, the addition of BPW caused an increase in thermal stability of the foam, for PUB presented a 375°C T_{onset} . This effect may be a result of greater interaction between hydroxyl groups of the lignocellulosic components found in the BPW and urethane groups, or its derivatives, which are present in the polymeric structure.

ATR-FTIR spectra shows that PUB foam presents bands characteristic of polyurethanes: the presence of NH linkage at 1621 cm^{-1} ; C=O stretching at 1701 cm^{-1} , C-O stretching at 1537 cm^{-1} and NCOO group at 1042 cm^{-1} . It could also be observed that the band's wide base between 3600-3200 cm^{-1} can be associated to the overlap of the bands related to NH and OH linkages. The absence of bands in the 2000-2300 cm^{-1} region confirms that all isocyanate groups have been involved in the reaction of foam formation.

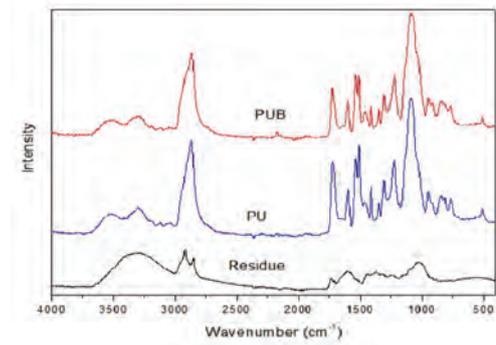


Figure 2 -ATR-FTIR spectra of the BPW, PU e PUB.

Fig. 3 shows SEM micrographs of PU and PUB foams, which show an irregular and macro-porous structure. It was possible to observe that PU consists of compacted channels, whereas PUB is formed by spheroid regions, with pores of smaller diameter – in comparison to PU – which intercommunicate between them. Such morphological differences can lead to different behaviors in the properties of adsorption and diffusion on the inside of the foams. Matins et al. (2015) also noted irregular surface and the porous morphology of BPW, what facilitated the adsorption of metals in aqueous solution.

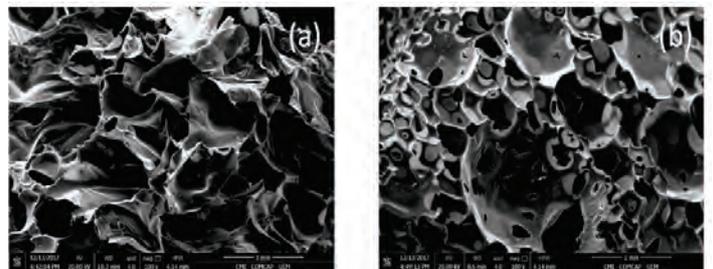


Figure 3 - Scanning electron microscopy of (a) PU foam and (b) foam containing banana peel (PUB). Magnification of 100 x.

ZERO POINT OF CHARGE (pH_{zpc}) / INFLUENCE OF pH ON ADSORPTION

The UV-Vis spectra of TAR solutions in the pH range of 3.0 to 8.0 presented the same maximum wavelength of absorption at 428 nm, thus evidencing that in such pH range there was the same maximum wavelength of absorption at 428 nm, thus evidencing that in such pH range there was the same protolytic structure of the dye or that the protolytic species absorb in the same wavelength.

Sulfonic groups, SO_3^- , carboxylate, COO^- , and azo, $\text{N}=\text{N}$, found in the TAR structure, show pK_a equivalent to 2.0, 5.0 and 10.86, respectively. Hence, the TAR molecule exists in its anionic form when the pH solution is greater than pK_a equal to 2, and in its neutral form whenever the solution pH is lower than pK_a equal to 2 (GOSCIANSKA AND PIETRZAK, 2015).

It was evaluated the influence of pH on adsorption by using a 35 mg L^{-1} aqueous TAR solution because this concentration result in values of absorbance that lead to good relation signal/noise for the spectrophotometric method and also valid by the Lambert-Beer law. From our experiments, highest amount of dye adsorbed was found at pH 3.0. The results of pH_{ZPC} for the foams, the pH considered optimal when it comes to TAR adsorption and its removal rate, and the removal rate at the natural pH value of the dye solution (pH= 6.0) are displayed by Table 2:

Adsorbent	pH_{ZPC}	% R (pH = 3.0)	% R (pH = 6.0)
PU	5.3 - 5.6	99.7	45.8
PUB	5.1 - 5.5	87.3	41.7

PU - foam without banana peel; PUB - foam containing banana peel.

Banerjee and Chattopadhyaya (2017) carried out the adsorption of TAR by using sawdust as an adsorbent, and they also reached maximum adsorption at pH 3.0. Nevertheless, they managed to achieve only 71% of dye removal, which was lower than the maximum obtained in this work by PUB (87.3%). Reck et al. (2018) tested TAR adsorption by charcoal activated by *babaçu* coconut and *moringa* seeds, and they found the pH optimal adsorption of 3.0 for both adsorbents. TAR adsorption by nano-molecularly imprinted polymer increased as pH was reduced from 10 to 2 (ARABZADEH et al., 2014).

The zero point of charge (pH_{ZPC}) is useful to predict superficial electrical charge in the adsorbent surface in relation to pH. Anion adsorption is favored when the pH value of the surface is lesser the

adsorbent's pH_{ZPC} value, for its surface will be positively charged. As for the adsorption of cations, it is favored whenever pH is higher than pH_{ZPC} , for the adsorbent surface will be negatively charged. pH_{ZPC} corresponds to the range in which the final pH remains constant, regardless of the initial pH value, that is when the material behaves as a buffer solution (GUILARDUCI et al., 2006).

PU and PUB pH_{ZPC} values are higher than the optimal adsorption of pH 3.0. Thus, their surfaces are positively charged at this pH, whereas TAR is found in its anionic form at this pH value, due to the predominance of SO_3^- and COO^- in the range of pH 2.0 and 5.0. Therefore, adsorption takes place between the surface of the positively charged foam and the anionic specie of TAR. At pH 6.0, the foams are negatively charged, and the TAR is also in its anionic specie which causes lesser adsorption at this pH value.

Kinetic study of adsorption

Evaluation of adsorption kinetics of TAR by PU and PUB foams demonstrated that, both at pH 3.0 and pH 6.0, the greatest q_e values were achieved in 120 min (Fig. 4). It can be observed that q_e values are similar between 60 and 120 min and that, after such period, they remained approximately constant. By using of *moringa* seeds for TAR adsorption at pH 5.0, Recket al. (2018) verified that the maximum adsorption was achieved in 60 min, and that the balance was spotted in 240 min.

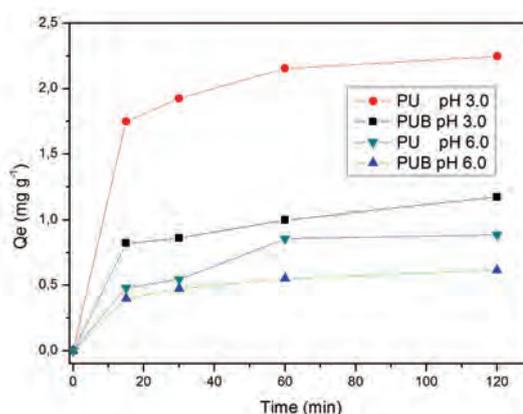


Figure 4 – Adsorption rate of TAR onto PU and PUB foams at pH 3.0 and pH 6.0, at 25.0 °C.

Analysis of the kinetic data of Q_e versus t showed that the pseudo-second-order model was the one to have the best adjustment to the experimental points for it shows R^2 values very close to the unit, as displayed by Figure 5.

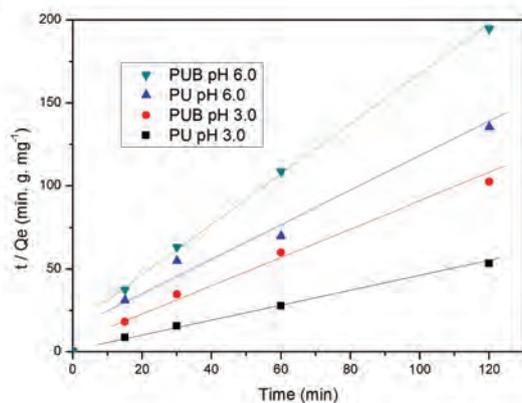


Figure 5 - Adjustment of the adsorption pseudo-second-order kinetic model for PU and PUB foams at pH 3.0 and 6.0.

The kinetic parameters are presented in Table 3. The maximum adsorption capacity achieved by pseudo-second-order (Q_e) is very close to the adsorption values ($Q_{t_{120}}$) experimentally reached. The kinetic model of pseudo-second-order indicates that adsorption is controlled by chemisorption, involving electrons donation or exchange between adsorbate and adsorbent. In this type of adsorption, molecules are not attracted to all the surface points, but specifically to the active centers, in a way to create a single layer (SILVEIRA et al., 2011). The kinetic study in which a chitosan and polyaniline composite was used also showed that TAR adsorption is controlled by the pseudo-second-order model (SAHNOUN AND BOUTAHALA, 2018). In other study that evaluated the adsorption by polyurethane foams of Procyon red HE-7B and methylene blue dye, the pseudo-second-order model was the one that best adjusted to the experimental data for both dyes (GÓES et al., 2016).

The results are in accordance with the optimal pH achieved for TAR adsorption, which was lower than pH_{PZC} value for both types of foam, thus confirming that the electrostatic attraction between the foam surface

and the charged dye is a limiting factor in the adsorption process.

TABLE 3 - Evaluation of kinetic parameters of the pseudo-second-order model for TAR adsorption by PU e PUB foams at pH 3.0 and pH 6.0, at 25°C.

Parameter	pH 3.0		pH 6.0	
	PU	PUB	PU	PUB
$Q_{t_{15}}$	1.749	0.823	0.481	0.400
$Q_{t_{30}}$	1.925	0.861	0.546	0.474
$Q_{t_{60}}$	2.156	0.999	0.857	0.552
$Q_{t_{120}}$	2.248	1.171	0.886	0.616
R^2	0.9997	0.9904	0.9748	0.9988
Q_e ($mg\ g^{-1}$)	2.357	1.270	1.049	0.673
k_2 ($g\ mg^{-1}\ min^{-1}$)	0.0726	0.0644	0.0472	0.1266

PU - foam without banana peel; PUB - foam containing banana peel.

CONCLUSION

The replacement of 10% (w/w) of polyethylene-glycol by BPW showed interesting results to foam production in relation to the chemical properties evaluated. The best results for TAR adsorption were achieved at pH 3.0, and the kinetic study showed that the greatest Q_e values were achieved in 120 min for both types of foam. The experimental data best fit the pseudo-second-order model, thus suggesting adsorption controlled by chemisorption. Despite PUB reached quite lesser value of TAR adsorption (87.3%) when compared to PU (100%), the results achieved by this study highlight an alternative to foam synthesis by making use of a renewable waste with low cost, that had adsorptive properties that can be useful when it comes to the decontamination of hydric bodies and the treatment of effluents, thus respecting environmental conservation.

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