Development of a Process for Color Improvement of Low-Grade Dark Maple Syrup by Adsorption on Activated Carbon

Amara Aït-Aissa, Natela Gerliani, Tatiana Orlova, Bita Sadeghi-Tabatabai, and Mohammed Aider*

ABSTRACT: Low-grade dark maple syrup was successfully discolored on activated carbon. Several experimental parameters were tested, namely, the mixing time (20, 40, and 60 min), concentration of the activated carbon (0.1, 0.3, and 0.5 g/100 mL), type of activated carbon (I, II, and III), activated carbon particle size (25, 50, and 75 μm), stirring speed (200, 400, and 600 rpm), and temperature (40, 60, and 80 °C). The obtained results showed that the discoloration is optimal by applying the following parameters: a mixing time of 40 min with a type III activated carbon at a concentration of 0.3 g/100 mL. These parameters yielded a light transmittance at 560 nm of 83.70 ± 0.21%, which ranks the syrup in the extra clear class according to the Canadian classification. The results showed that among the tested carbons, the adsorption on the type III carbon followed the Langmuir, Freundlich, and Langmuir−Freundlich adsorption isotherms. Regarding the effect of the particle size, the obtained results showed that a mean size of 25 μm combined with a stirring speed of 200 rpm and working temperature of 80 °C was the most effective one. The optimized conditions showed a good adequacy with the Langmuir and Freundlich models. The discoloration process by using the type III activated carbon followed the pseudo-second-order kinetics.

1. INTRODUCTION

Maple syrup is largely produced in North America by heat evaporation of maple sap collected from maple sugar trees (Acer saccharum) during the early spring season. According to Statistics Canada 2019, the Canadian maple syrup industry accounts for approximately 71% of the world’s maple syrup production. As such, Canada is the leading global producer of maple products, with 91% originating from within Quebec, 5% from Ontario, and 3% from New Brunswick. The United States is the second largest world producer accounting for approximately 27% of global production, with 47% originating from Vermont, 19% from New York, and 13% from Maine (Statistics Canada, 2019). In Canada, the maple syrup is mainly classified according to the color by measuring its light transmittance at 560 nm, which is the most convenient wavelength to distinguish syrups with high precision according to their ability to absorb-transmit visible light. There are two types of classification for maple syrup: the federal government classification and that of the provincial government. The Canadian Food Inspection Agency governs the quality and safety of maple products in Canada and is responsible for the federal classification of maple syrup, which can be classified as extra light, light, medium, amber, and dark. The latter is generally associated with a strong burnt caramel flavor and taste. Moreover, huge quantities of maple syrup called “unclassified” are produced each year. This product is stored and large quantities are accumulated from year to year, creating serious problems with management and marketing. Indeed, gradually as the season progresses, the fructose and glucose content increases in the sap, while the sucrose content decreases. Moreover, the content of other natural compounds found in the sap is also changing during the season such as amino acids and minerals. These changes in the composition of...
the sap affect the color and flavor of the maple syrup, mainly because of the Maillard reactions that occurred between the reducing sugars and amino acids of the sap during the heat evaporation process. Early in the season, the syrup is generally clear with good sweet taste, corresponding to the syrup of the best quality. However, as the season progresses, the syrup becomes darker and tastes like caramelized sugar with less refined flavor.

To increase the profitability of the maple syrup industry, it is important to commercialize all the produced products, including the dark syrup called unclassified. To achieve this objective, it is necessary to improve the color of the syrup and make it amber or light. Moreover, the materials and processes used to improve the color of the syrup must positively affect the sensory quality of the end product, namely, the color, typical smell, flavor, and taste of maple syrup. Since maple syrup is a food product, the used material for color correction must be safe and economically affordable. In this context, activated carbon (charcoal) seems to be an appropriate decolorizing and taste/smell-correcting (improving) agent. However, the choice of the most appropriate type of activated carbon for a particular application must be technologically feasible because the achievement of the targeted objectives will depend on different factors such as the physical and chemical properties of the adsorbed material, intrinsic functional properties of the activated carbon, and other experimental conditions that can play an important role in the adsorption process.

At the end of the 18th century, the adsorption properties of activated carbon were observed. Then, the activated carbon was used for the first time in England in the sugar industry in 1794 to improve the process efficiency and product quality in the white sugar-making industry. However, the modern industrial production and the use of activated carbon were reported for the first time in a patent deposed by Ostrejko in 1901. Today, activated carbon is used in several industrial applications, including gas and air cleaning, purification and recovery of different materials and substances, environmental protection, and the removal of hydrocarbons and solvents from chemical reactions. Furthermore, the activated carbon is also increasingly used in the treatment of water, including drinking water, groundwater, and wastewaters. Its main role is the absorption of dissolved organic/inorganic impurities and the elimination of all substances that affect the smell, taste, and color of the treated materials. Furthermore, the activated carbon is widely applied for liquid discoloration, which is particularly important in the pharmaceutical and food industry, including the maple syrup industry.

This paper reports an experimental work with a subsequent theoretical interpretation of the mechanisms involved in the dark maple syrup discoloration by activated carbon. The goal of this study was to use a food-grade activated carbon as an adsorption material to improve the color and smell/taste of dark maple syrup in order to produce lighter syrup corresponding to syrup of light or at least amber grade. Specifically, the effect of many experimental factors such as the agitation time ($X_1$), activated carbon mass in the sample ($X_2$), and activated carbon type ($X_3$) on the syrup light transmittance was studied by using a three-level full factorial experimental design. The kinetics of adsorption of the maple syrup pigments were interpreted (discussed) by using two kinetic models: the pseudo-first-order and pseudo-second-order kinetic models. Kinetic parameters and correlation coefficients were also determined. Moreover, the adsorption isotherm models of Langmuir, Freundlich and Langmuir–Freundlich were used to evaluate the effect of the experimental parameters on the performance of the activated carbon to adsorb the maple syrup pigments responsible for the dark color.

2. MATERIALS AND METHODS

2.1. Reagents and Samples. Dark, low-grade (unclassified) maple syrup was purchased from a local maple syrup farm in Quebec City, Canada. The used maple syrup was
characterized by a Brix degree of 65 ± 1 °Brix. The type I, II, and IV activated carbons were from Sigma-Aldrich (St. Louis, MO, USA). The type III activated carbon was obtained from Fisher Scientific (Waltham, MA, USA).

2.2 Characterization of the Activated Carbon.
2.2.1 Mean Particle Size. The mean particle size of each activated carbon used in this study was determined by the sieving method by using a series of sieves from the Canadian Standard Sieve Series (W.S. Tyler Company, ON, Canada) according to the ASTM procedure.14

2.2.2 Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray Spectrometry (EDS). Scanning electron microscopy (SEM) images of the activated carbons were taken by using an electron microscope (Joel, JSM-840A, North Billerica, MA, USA) equipped with an energy dispersive X-ray spectrometer (PGT Instrument, model Avalon, Princeton, NJ, USA). The energy dispersive X-ray spectrometry (EDS) condition was set at 15 kV. The samples were first metalized by coating with a thin gold/palladium layer to make the surface highly conductive and allow the free flow of the excess electrons. This procedure is necessary to prevent the sample to be charged when it is exposed to the SEM electron probe.15

2.2.3 Activated Carbon Bulk Density. The bulk density of the activated carbon used in this work is calculated by the tube method as adapted from the ASTM method D2854-09.16 A known amount of each activated carbon was weighed and poured into a test tube, which was tapped with 10 strokes. Then, the resulting volume of the activated carbon in the sample was read. Knowing the mass and volume, the bulk (apparent) density of each activated carbon was determined.

2.3 Experimental Procedure. The discoloration process was carried out by using two syringe filters with mesh sizes of 45 and 20 μm, respectively. Finally, the light transmittance of the recovered samples was analyzed by using a UV–Visible spectrophotometer (Cary 60 UV–Vis, Santa Clara, CA, USA) at a wavelength of 560 nm at ambient temperature of 22 ± 1 °C.17

2.4 Experimental Design and Statistical Analysis. In the first experimental block, a full factorial experimental design (3³) was used and all the experiments were carried out at ambient temperature (~25 °C). The independent variables were as follows: agitation time (X₁) (20, 40, and 60 min), activated carbon concentration (X₂) (0.1, 0.3, and 0.5%), and activated carbon type (X₃) (I, II, and III). The second experimental block was also carried out as a full factorial design with three independent variables: working temperature (X₁) (40, 60, and 80 °C), activated carbon mean particle size (X₂) (25, 50, and 75 μm), and agitation speed (X₃) (200, 400, and 600 rpm). The analysis of variance (ANOVA) and normality test (Shapiro–Wilk) were used to investigate the differences between the mean values of the compared treatments at a 95% significant level by using SigmaPlot v.11 software (Systat Software Inc., San Jose, CA, USA) and Maple Software v.14 (Maplesoft, Waterloo, ON, Canada). The constants of the adsorption isotherm models (Langmuir, Freundlich, and Langmuir–Freundlich) (Table 1), average relative errors (%), and coefficients of determination (R²) based on the actual deviation between the experimental points and predicted values were estimated by using SigmaPlot v.11 (Systat Software, Inc., San Jose, CA, USA). All experiments were carried out at least in triplicate and mean values ± SD were used for different calculations and comparisons.

3. RESULTS AND DISCUSSION

3.1 Characteristics of the Used Activated Carbons. The mean particle size analysis served to determine the size distribution of the particles constituting the different activated carbons used. The total carbon mass, which was sifted, was 200 g and the particle size distribution was lower than 300 μm. The obtained results showed that the particle size distribution was dependent on the form of the carbon (powder or grain) and on the particle shape (spherical or heterogeneous shape). It has been found that the used carbons did not present a uniform size distribution and have heterogeneous shapes including a spherical form. Moreover, the type III activated carbon has a particle size diameter lower than 150 μm (Figure 2).

Figure 2. Activated carbon particle size distribution.

The different activated carbons used in this work were inert carbonaceous materials. Some of them have highly developed intrinsic porosity, which gives them good adsorbing properties, facilitating the fixation of the colored pigments present in the dark maple syrup on their surface.18 This feature is due to the micropores present in these carbons. The number and distribution of these micropores can significantly affect the

Table 1. Adsorption Isotherm Models Used for Color Improvement of Maple Syrup

| model            | equation                                                                 | linear equation |
|------------------|--------------------------------------------------------------------------|-----------------|
| Langmuir         | qₑ = \frac{Q_K C_{ₑ}}{1 + \frac{Q_K C_{ₑ}}{K_L}}                         | \frac{1}{qₑ} = \frac{1}{Q_K} + \frac{1}{C_{ₑ}}          |
| Freundlich       | qₑ = K_n C_{ₑ}^n                                                       | ln(qₑ) = ln(K_n) - \frac{n}{n} ln(C_{ₑ})                 |
| Langmuir–Freundlich | qₑ = \frac{Q_K (K_n C_{ₑ})^{n-1}}{1 + \frac{Q_K C_{ₑ}}{K_L}}            | \frac{1}{qₑ} = \frac{1}{Q_K} + \frac{1}{Q_K C_{ₑ}^{1/n}} + \frac{1}{C_{ₑ}} |

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adsorption capacity of these materials. Unlike the type IV activated carbon, the micropore distribution was much pronounced on the surfaces of type I, II, and III activated carbons (Figure 3). The structure of type III activated carbon seems to be highly developed than the other carbon types. The pore structure increases the specific surface area of the activated carbon, which can reach approximate values of 1,500 m²/g carbon.¹⁰ This specific surface area increases the carbon adsorption properties. For the texture, the type I, II, and IV activated carbons have an amorphous texture, which is made of graphite microcrystals in different interconnected forms. The type III activated carbon has a crystalline structure and specific
sites, which are favorable for good adsorption properties. In addition, each microcrystal comprises a stack of several crystalline layers with a high degree of porosity.

The structural analysis showed that they are mainly consisted of carbon with various types of elements in different amounts. Besides carbon, the type I activated carbon contains some volatile matter, in particular oxygen. The other composition is represented by ash material composed of some volatile matter, in particular oxygen. The other amounts. Besides carbon, the type I activated carbon contains punctual interparticle contact (Figure 5).

The results obtained for the bulk density showed that the values corresponding to the type I, II, and III activated carbons are almost similar. These carbons are used in the form of a powder to discolor the dark maple syrup. The type IV activated carbon was different from the other carbons and was characterized by a high bulk density, which may be due to its initial granular form. In addition, the observed differences in the bulk densities can be attributed to the differences observed in the particle shape of the used activated carbons. Applied to the targeted adsorption application of this study, the spherical form of the particles of the activated carbon will be more favorable because this form has a low bulk density with a punctual interparticle contact (Figure 5).

Figure 5. Bulk density comparison of the different activated carbons.

3.2. Effect of the Concentration and Type of the Added Activated Carbon. The evolution of the maple syrup light transmittance measured at 560 nm as a function of the mixing time and different concentrations of the activated carbon added is shown in Figure 6. Statistical analysis of the obtained data showed a significant effect ($p < 0.001$) of the mixing time. The plotted values of the dependent variable (light transmittance) followed a behavior that can be described by a second-order polynomial equation. It has been observed that the light transmittance of the treated maple syrup increased by increasing the concentration of the activated carbon added ($p < 0.001$). This can be explained by the increase in the contact surface between the adsorbent (activated carbon) and adsorbate (dark maple syrup), which yielded a high amount of adsorbed colored pigments. In fact, it has been already reported that the higher the contact area is, the higher the absorption reaction is. In Figure 6a, which corresponds to the data obtained by using the type I activated carbon, it can be seen that the light transmittance increased from 11.11 ± 0.25% up to 31.39 ± 0.30% in the case of using 0.1% of activated carbon and up to 64 ± 0.3% in the case of using 0.3%. However, addition of 0.5% of activated carbon resulted in final syrup with a light transmittance of 56 ± 0.32%, which is lower than the value obtained with 0.3%. On the other hand, Figure 6b shows that the light transmission varied between 11 and 17% in the case of using the type II activated carbon. At the end of the mixing time of 60 min, values of the light transmittance of the maple syrup were 15.11 ± 0.25, 15.65 ± 0.3, and 16.63 ± 0.25% for the activated carbon concentrations of 0.1, 0.3, and 0.5%, respectively. This result showed the nonsignificant effect ($p > 0.05$) of the concentration of type II activated carbon. Finally, Figure 6c shows that the light transmittance varied between 23.42 and 61.51% in the case of using the type III activated carbon. The final light transmittances of the obtained syrup after 60 min treatment were 23.42 ± 0.31, 83.86 ± 1.15, and 61.51 ± 0.51% for a concentration of the added type III activated carbon of 0.1, 0.3, and 0.5%, respectively. This type of activated carbon (type III) gave the highest light transmittance of the final maple syrup, which was probably due to its large surface area. The results obtained with the type IV activated carbon (Figure 6d) were not significantly different from those obtained with the type II activated carbon. The optimized results showed that the optimal activated carbon concentration to be added to the dark maple syrup to obtain a final product with a significantly improved color is 0.3%. Thus, the use of 0.5% of activated carbon is not necessary since the adsorption phenomenon that occurred was the highest (saturated) at 0.3%.

This saturation can be explained by the chemisorption process, which is more dominant in the present case. Indeed, it has been reported that chemisorption is a common process in the adsorption of dyes on different adsorbents. Finally, it has been found that the use of type I activated carbon gives a syrup that can be classified as amber with a light transmittance.
ranging between 27 and 43.9%. The type II and IV activated carbons did not improve the color of the used dark maple syrup. The use of type III activated carbon did not improve the color of the syrup when it was used in a concentration of 0.1%, but it gave a syrup that can be classified as light when this activated carbon concentration was 0.5% or extra light when its concentration was 0.3%. Thus, the highest light transmittance of the obtained syrup can be obtained with the type III activated carbon at a concentration of 0.3% in the working mixture.

The evolution of the light transmittance of the treated maple syrup as a function of the activated carbon type is shown in Figure 7a–c. It can be observed that the light transmittance of the treated syrup was dependent on the activated carbon type and increased by increasing the concentration of the used activated carbon. The light transmittance was the highest when type III carbon was used. It was followed by the type I and II carbons. The type IV activated carbon gave the worst results in terms of improving the light transmittance of the syrup. In all cases, the obtained light transmittances after 20 and 40 min of mixing by using the type III activated carbon are higher than the one obtained with the type I activated carbon even after 60 min of mixing. This can be explained by the fact that the colored pigments are more adsorbed by using the type III activated carbon.

3.3. Effect of the Mixing Time at Different Activated Carbon Concentrations. The evolution of the light transmittance of the maple syrup as a function of the mixing time (20, 40, and 60 min) with different activated carbon types is shown in Figure 8a–d. Figure 8a shows that by using the type I activated carbon, the effect of the mixing time on the adsorption of the colored pigments was not significant (p > 0.05). The highest light transmittance recorded with a concentration of 0.1% of type I activated carbon was 37.82 ± 0.12% after 40 min. With a concentration of 0.3% of the activated carbon, the light transmittance reached a value of 64 ± 0.11% after 60 min, and finally, by using a concentration of 0.5% of the activated carbon, a light transmittance of 68.02 ± 0.12% after 40 min of mixing was obtained. Figure 8b shows that the type II activated carbon differently adsorbs the colored pigments compared to the type I activated carbon as a function of the mixing time for a given (fixed) activated carbon concentration. Indeed, by using a concentration of 0.1 and 0.3% of activated carbon, the light transmittance increased linearly as a function of the mixing time. At the same time, the obtained results showed that at a concentration of 0.5% of the activated carbon, the effect of the mixing time on the color of the treated maple syrup was not significant (p > 0.05). In Figure 8c, which corresponds to the use of type III activated carbon, the obtained results show that the adsorption kinetics reached a maximum by using a treatment time of 40 min. In this case, a maximum light transmittance of 83.70 ± 0.21% was obtained. Beyond this time, the adsorption was saturated. However, at a fixed concentration of the used activated carbon, the effect of the mixing time was the lowest for a 20 min treatment. No significant difference was observed between 40 and 60 min of mixing.

The adsorption of the colored pigments on the surface of the activated carbon is achieved in four steps,26,27 which can be described as follows: The first step represents the migration of the coloring pigments from the syrup to the surface of the activated carbon. The second one is the diffusion of the colored pigments through the interparticle spaces, which corresponds to the external diffusion phase. The third step is the intraparticle diffusion, and the last one represents the surface chemical reaction between the activated carbon surface (adsorbent) and the active groups of the maple syrup pigments. It should be noted that the first step can be controlled with an appropriate mixing time, while the last step is very fast, suggesting that the diffusion processes are the limiting steps that control the adsorption phenomenon.26

3.4. Effect of the Temperature, Carbon Particle Size, and Mixing Speed. The effect of the temperature (40, 60, and 80 °C) on the adsorption of the pigments from the dark maple syrup by using the type III activated carbon is shown in Figure 9. The increase in the temperature resulted in a significant (p < 0.001) increase in the light transmittance of the final maple syrup. This increase in the temperature caused an
increase in the adsorption reaction by increasing the average velocity of the molecules. As a result, the molecules of the colored pigments acquired sufficient kinetic energy to produce effective collisions with the adsorption surface of the activated carbon.\textsuperscript{29−31} The effect of the particle size was also significant ($p < 0.001$) and Figure 9a–c shows that the increase in the light transmittance varies according to the particles of the used activated carbon type. By using the particle size of 25 $\mu$m, the highest light transmittance reached an average value of 78.37 ± 0.15%. For the same type of the activated carbon (type III), the use of particle sizes of 50 and 75 $\mu$m yielded average light transmittances of 67.48 ± 0.10 and 62 ± 0.35%, respectively. Thus, it seems that the adsorption phenomenon increases by increasing the specific surface area of the activated carbon, which corresponds to the lowest particle size.\textsuperscript{32}

The effect of the particle size of type III activated carbon on the adsorption of the pigments from the dark maple syrup is shown in Figure 10a–c. The data plotted show a maximum discoloration of the maple syrup, which corresponds to a light transmittance of 68 ± 0.10%. This result was obtained under the following experimental conditions: mean particle size of 25 $\mu$m, working temperature of 80 °C, and stirring speed of 200 rpm. Moreover, Figure 10c shows two interesting trends. The first was up to 200 rpm with a rapid rise of the light transmittance, followed by the formation of a steady state between 200 and 600 rpm.

The effect of the mixing (agitation) speed on the adsorption of maple syrup coloring pigments on the surface of type III activated carbon is presented in Figure 11, on which it can be seen that the light transmittance of the treated syrup is inversely proportional to the increase in the mixing speed. This can be explained by the fact that the increase in the mixing speed caused desorption of some already adsorbed colored pigments from the surface of the activated carbon. The optimized results showed an optimum stirring speed of 200 rpm, which can be considered as sufficient to promote an adequate contact between the activated carbon particles and the colored pigments (molecules) of the dark maple syrup.\textsuperscript{33}

3.5. Adsorption Isotherm Models. Figure 12 shows the application of adsorption isotherm models to predict the discoloration process of the maple syrup under different experimental conditions. This discoloration process was considered to be a result of adsorption phenomena. The experimental results were analyzed by using three two-parameter isotherm models: Freundlich, Langmuir, and three-parameter Langmuir–Freundlich isotherm model. The main characteristic of the Langmuir adsorption isotherm is its simplicity and the physical meaning of the $K_L$ and $Q_m$ parameters. The $K_L$ is related to the strength of the interaction between the adsorbed molecules and the solid surface, while the $Q_m$ value expresses the amount of a solute per gram of a solid surface, which is considered as totally covered by a monomolecular layer. This model is verified by a monolayer
adsorption model and demonstrates the heterogeneity of the surface. Regarding the use of type I activated carbon, Figure 12a shows that the experimental points do not fit the Langmuir adsorption isotherm model. This result can be explained by the absence of interactions between the adsorbed species and the adjacent surface of the activated carbon. Similarly, the difference could come from either significant lateral interactions or a distribution of more complicated sites. In the case where the type II and III activated carbons were used, Figure 12b,c shows that the experimental results are verified by the Langmuir adsorption isotherm model. Unlike the type I activated carbon, the type II and III activated carbons provide an adsorbed monolayer of the colored pigments contained in the used dark maple syrup. This can be explained by the fact that the type II and III activated carbons have heterogeneous surfaces and that all their adsorption sites are energetically equivalent. Moreover, the obtained coefficients of determination are 0.84, 0.96, and 0.99 for the type I, II, and III activated carbons, respectively. The calculation of the quantities of the colored pigments that are fixed on the activated carbon surface, which is considered as totally covered by a monomolecular layer, varies depending on the type of the used activated carbon (I, II, and III). These quantities are 0.72 $\times 10^{-5}$, 2.64 $\times 10^{-5}$, and 3.89 $\times 10^{-5}$ g/g of the type I, II, and III activated carbons, respectively. The obtained results were also verified if they fit the Freundlich adsorption isotherm model. This choice is justified by the fact that this adsorption isotherm model is linked to a constant indicating the adsorption capacity of the activated carbon, as well as to an empirical constant, which is related to the magnitude of the driving force of the adsorption phenomenon. The use of type I and II activated carbons (Figure 12a,b) shows a weak adequacy of the Freundlich adsorption isotherm model with the experimental results. This can be explained by the fact that the coloring pigments (molecules) of the dark maple syrup do not necessarily follow a multilayer adsorption behavior. In the case when the type III activated carbon was used (Figure 12c), the experimental results adequately fitted the Freundlich adsorption isotherm model. The coefficients of determination ($R^2$) were 0.73 and 0.78 in the case of the use of type I and II activated carbons, respectively, and 0.97 in the case when the type III activated carbon was used. Also, comparing the constants that serve to indicate the adsorption capacity of the adsorbent, the one (constant) related to the type III activated carbon ($K_f = 1.95$ mg L$^{-1}$ g$^{-1}$) is significantly higher than those related to type I and II activated carbons, which are 0.98 and 0.99 mg L$^{-1}$ g$^{-1}$, respectively.

Furthermore, Figure 12c shows that the experimental results adequately fit all the Langmuir, Freundlich, and Langmuir–Freundlich adsorption isotherm models when the type III activated carbon was used to discolor the dark maple syrup. This type of activated carbon (type III) adsorbs the molecules of the colored pigments by monolayer and multilayer adsorption mechanisms. Sips showed that the monolayer and multilayer adsorption mechanisms are both complex phenomena because there is superposition of saturation adsorption energy on the active sites along a curved homogeneous Langmuir adsorption isotherm model and on heterogeneous sites according to the Freundlich isotherm adsorption model.34 This superposition of different phenomena can be interpreted in terms of the distribution of the coloring pigments on the surface of the used activated carbon. Some pigments diffuse into the activated carbon and the concentration of this fraction follows Freundlich’s law. Another fraction of the colored pigments will be adsorbed on the surface of activated carbon, and this process will be adequately described by the Langmuir adsorption isotherm model. The coefficients of determination obtained in Figure 12c for the Langmuir, Freundlich, and Langmuir–Freundlich adsorption isotherm models by using the type III activated carbon are 0.99, 0.98, and 0.99, respectively. These results indicate that characteristics of the type III activated carbon are the most suitable to achieve the main objective of this work, which consists of removing coloring pigments from the used dark maple syrup to improve its quality by increasing its light transmittance ability in the visible spectrum at 560 nm.

3.6. Kinetic Order of the Discoloration of the Dark Maple Syrup. The kinetic orders of the adsorption of the colored pigments of the dark maple syrup on the type III activated carbon are given in Table 2 and graphically shown in Figure 13.

![Figure 12. Coloration concentration of the maple syrup solution as a function of coloration concentration adsorbed by activated carbons of different adsorption isotherm types: (a) type I activated carbon, (b) type II activated carbon, and (c) type III activated carbon.](https://dx.doi.org/10.1021/acsomega.0c02717)
treatment. This step can be characterized by a balance between the adsorbed molecules on the surface of the activated carbon and the desorbed ones. The major part of the coloring molecules, which were transferred to the surface of type III activated carbon, was observed in the first 40 min of the treatment. This was confirmed by the experimentally obtained light transmittance of 83.7 ± 0.2%. In addition, the experimental results do not fit the first-order kinetic model. These observations led us to conclude that the adsorption of the colored pigments of the dark maple syrup by using the type III activated carbon is not a controlled diffusion process since it does not follow the first-order equation given by Corbett.\(^\text{35}\) Figure 13b shows the application of a pseudo-second-order kinetic model to describe the results obtained for the adsorption of the colored pigments of the dark maple syrup on the type III activated carbon. The plot of \((t/q_t)\) versus time \((t)\) allowed us to determine by extrapolation the constant of the pseudo-second-order kinetic model at different contact times between the syrup and used activated carbon. The value of \(k_2\) obtained is 0.0049 L mg\(^{-1}\) min\(^{-1}\). In view of these results, it appears that the amount of the adsorbed molecules increases by increasing the mixing time. Furthermore, the experimental results adequately fit the pseudo-second-order kinetic. Thus, the result leads us to conclude that the adsorption process followed the model that can be described by the pseudo-second-order kinetics.

4. CONCLUSIONS

In this work, different activated carbons were used to improve the color of a dark maple syrup. Scanning electron microscopy (SEM) analysis of the used activated carbons showed that they are different from each other by their specific and contact surface areas, as well as by their roughness. Moreover, energy dispersive X-ray spectrometry (EDS) showed that the chemical composition of these activated carbons is also different from each other.

By using the activated carbon defined as type III carbon, the adsorption of the coloring pigments from the dark maple syrup led to high elimination of the dark color and the process yielded a syrup with a light transmittance of 83.70 ± 0.2%. This syrup can be classified as extra clear according the Canadian classification of maple syrup. This result was obtained under the following operating conditions: type III activated carbon used at a concentration of 0.3% at a mixing time of 40 min. The optimized experimental conditions showed that the adsorption of the coloring pigments on the surface of the activated carbon was favored by a particle size of 25 μm under an operating temperature of 80 °C and mixing speed of 200 rpm.

The adsorption kinetics of the discoloration process of the dark maple syrup under different experimental parameters was verified according to different adsorption isotherm models. Two-parameter Langmuir and Freundlich isotherm models, as well as a three-parameter Langmuir–Freundlich isotherm model, were tested. It has been shown that the discoloration of the dark maple syrup by using the type III activated carbon can be predicted by the Langmuir, Freundlich, and Langmuir–Freundlich isotherm models, depending on the experimental conditions used. Unlike the pseudo-first-order kinetics, it has been shown that the discoloration of the dark maple syrup by using the type III activated carbon can be described by the pseudo-second-order kinetics.

**AUTHOR INFORMATION**

**Corresponding Author**

Mohammed Aider — Department of Soil Sciences and Agro-Food Engineering and Institute of Nutrition and Functional Foods (INAF), Université Laval, Quebec, Quebec G1V 0A6, Canada; orcid.org/0000-0003-1487-9274; Phone: (418) 656-2131 # 409051; Email: mohammed.aider@fsaa.ulaval.ca

**Authors**

Amara Ait-Aissa — Department of Soil Sciences and Agro-Food Engineering, Université Laval, Quebec, Quebec G1V 0A6, Canada

Natela Gerliani — Department of Soil Sciences and Agro-Food Engineering and Institute of Nutrition and Functional Foods (INAF), Université Laval, Quebec, Quebec G1V 0A6, Canada

Tatiana Orlova — Kuban State Agrarian University, 350044 Krasnodar, Russian Federation

Bita Sadeghi-Tabatabai — Department of Soil Sciences and Agro-Food Engineering, Université Laval, Quebec, Quebec G1V 0A6, Canada

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