Orange-derived and lemon-derived adsorbents with controlled grain for an efficient elimination of some cationic and anionic dyes

ABSTRACT

Purpose
The study aims to demonstrate that orange-derived and lemon-derived systems can be used in continuous processes as efficient adsorbents to the entrapment of some anionic and cationic dyes in the textile dyeing wastewater effluents.

Design/methodology/approach
Physically and chemically modified orange and lemon mesocarps are used as natural adsorbents for the cationic dyes Basic Blue 3, Basic Yellow 21, Basic Red 18 and Basic Green 4 and, the anionic dyes Acid Blue 264, Acid Yellow 49 and Acid Red 337; all commonly used in the textile dyeing industry. Adsorption capacities of the orange-derived and lemon-derived adsorbents on the dyes are studied simulating a batch and continuous industrial processes.

Findings
Results demonstrate that treated orange mesocarp (orange-derived adsorbent) can adsorb up to 97% of cationic Basic Green 4 in 30 min while the lemon mesocarp (lemon-derived adsorbent) can retain up to 88% within the same time. In the case of anionic, 91% Acid Blue 264 is adsorbed by the orange mesocarp in 15 min whereas a 92% is adsorbed by the lemon homologue within the same time.

Originality/value
As far as we know, physically and chemically modified orange and lemon mesocarps have not been used on the removal of cationic (Basic Blue 3, Basic Yellow 21, Basic
Red 18 and Basic Green 4) and anionic (Acid Blue 264, Acid Yellow 49 and Acid Red 337) dyes of textile dyeing wastewater industry. It is a costless and efficient treatment that supposes, on the one hand, an ecofriendly and feasible process for discolouration of wastewater and, on the other, a valorisation (upcycling) of orange and lemon peels, which are not currently used.

**KEYWORDS:** cationic dye, anionic dye, adsorbent, adsorption, citrus peel, wastewater, textile dyeing

**Introduction**

One of the main drawbacks of the textile dyeing industry is the generation of large amounts of wastewater. This wastewater is rich in colour (dye) and in chemicals like surfactants, salts, alkalis and other hard-degradation and organic compounds that result in high-concentrated COD and BOD effluents (Hauser, 2011). The environmental impact includes solids in suspension, ionic charge, toxicity, oxygen concentration and colourisation. Generally, most dyes are very persistent in wastewaters due to their solubility and low degradation degree both determined by the complexity of their structure (Buitrón et al., 2004). It is estimated that approximately 100 tonnes/year of dyes are discharged into water and about 70% are azo type dyes (Gupta and Suhas, 2009; Ihsanullah et al., 2020). With the aim to discolour streams, several techniques have been investigated, including chemical (oxidation, electrolysis) (Corona-Bautista et al., 2021; Pacheco-Álvarez et al., 2019; Suhadolnik et al., 2019), ozonation (Abrile et al., 2020), physical (reverse osmosis, filtration, coagulation/flocculation, adsorption) (Ahmad et al., 2015; Al-Aoh, 2021; Paixão et al., 2020), combination of chemical and physical (Rezaee et al., 2008) and even biological (microorganisms, enzymes) processes (Arabaci and Usluoğlu, 2014; Garg and Tripathi, 2017; Saratale et al., 2011). The
majority of those operations incur in, among others, elevated maintenance level and manpower with high expertise that affect the final cost of the process and the profitability of the production (Donkadokula et al., 2020; Hauser, 2011; Soares et al., 2017).

Among all the water discoloration techniques, adsorption emerges as an economic alternative and a low environmental impact process (Liu et al., 2020; Rashed, 2013). The adsorption mechanism is based in the capture or retention of a compound (adsorbate) in a liquid phase by a solid (adsorbent) (Mostafa et al., 2009; Park et al., 2010). In this sense, a wide range of low-cost materials have been studied to be used as potential adsorbents for dyes such as wood (Kelm et al., 2019; Naeem and Hassan, 2018), clay (Kausar et al., 2018), ashes (Mor et al., 2018), activated muds (Qu et al., 2012; Sarvajith et al., 2018), orange (Namasivayam et al., 1996), cuttlefish bones (Elwakeel et al., 2020), algae (Elgarahy et al., 2020) and banana peels (Temesgen et al., 2018). In these recent years, the concepts of reuse and recycling have taken an important role in our society and resulted in an increase in research and development in sustainable technologies. For this reason, the current trend is to use agroindustrial wastes as alternative to produce new adsorbent materials due to their low cost and efficiency in the elimination of metal ions (Fu and Wang, 2011; Mo et al., 2018; Vijayaraghavan and Balasubramanian, 2015).

Part of the transformation of agroindustrial by-products such as orange and lemon peels into new raw materials, concretely into dye adsorbents, has been performed by CRESCA research group. In this sense, previous studies demonstrated satisfactory results in the use of mesocarps of orange and lemon into the adsorption of organic dyes as alternative for textile industry wastewater treatment (Arjona et al., 2018) or even as wine clarifiers (García-Raurich et al., 2019).
Here, the evaluation of the adsorption capacity of modified mesocarps of orange and lemon peels to retain cationic and anionic dyes from textile dyeing industry wastewaters is presented. The adsorption capacity of the resulting adsorbents is tested in batch and in continuous processes with different cationic and anionic dyes, simulating industrial operations. Parameters such as the nature of the adsorbent, the concentration of the adsorbent, pH of the adsorption process, time of contact, hydration degree and number of column pass are evaluated by analysing the concentration of the dye in the residual liquor fraction by UV-Vis spectroscopy.

**Materials and methods**

**Materials**

Orange and lemons peels were collected from local fruit stores. Cationic dyes: Basic Blue 3 (C_{20}H_{26}ClN_{3}O) (Blue), Basic Yellow 21 (C_{22}H_{25}ClN_{2}) (Yellow), Basic Red 18 (C_{10}H_{25}Cl_{2}N_{3}O_{2}) (Red) and Basic Green 4 (C_{23}H_{25}ClN_{2}) (Green). Anionic dyes: Acid Blue 264 (C_{29}H_{28}N_{3}NaO_{6}S_{2}) (Blue), Acid Yellow 49 (C_{18}H_{12}Cl_{2}N_{3}O_{3}S) (Yellow) and Acid Red 337 (C_{17}H_{11}F_{3}N_{3}NaO_{4}S) (Red) (**Fig. 1**). Dyes were purchased from Classic Dyestuffs Inc. Filters of 0.45 µm pore of Millex were used in the batch process.

**Fig. 1** a) Basic Blue 3, b) Basic Yellow 21, c) Basic Red 18, d) Basic Green 4, e) Acid Blue 264, f) Acid Yellow 49 and, g) Acid Red 337.

**Physical treatment of peels**

Washing of the orange and lemon peels was carried out with tap water and conventional soap at room temperature to eliminate added waxes, resins and soil of the epicarp. After drying with continuous air flow, mesocarps were cut using an ice mincer until achieve 0.5-1 µm particle sizes, according to previous studies (Arjona et al., 2018).
Chemical treatment of peels

Due to the different nature of the studied dyes, two different chemical treatments were applied to peels. With the objective to achieve an adsorbent capable to adsorb anionic dyes, the procedure was:

1. Acidic treatment: to remove pectins and other compounds such as β-carotene (50 g peel/500 mL deionised water, pH = 3-4, ultrasonic agitation, 45 min).
2. Alkaline treatment: saponification of peels, concretely cellulose, hemicellulose and remaining pectin fraction (50 g peel, 2.5 g Ca(OH)\(_2\) (Panreac), the necessary volume of deionised water, pH > 10, ultrasonic agitation, 45 min).
3. Acidic treatment: modification of ionic charge (50 g peel/500 mL deionised water, pH = 3-4, ultrasonic agitation, 45 min).
4. Neutralisation
5. Dry

The third step was avoided when peels were prepared to adsorb cationic dyes.

Batch process

In the discontinuous or batch process, the amount of adsorbent (0.50, 0.75 and 1.00 g), type of dye (anionic and cationic), concentration of dye (30, 60, 90, 120 and 240 ppm), pH (as-it-is, acid and alkaline), adsorption time (5, 10, 15, 30 and 45 min for anionic and 15, 30, 45, 60 and 75 min for cationic) and the initial hydration degree of orange and lemon adsorbent parameters were studied. To do this, 25 mL of a solution of dye was prepared and mixed with the corresponding adsorbent at 20ºC. Then, the sample was filtered and the concentration of dye in the liquor was analysed by UV-Vis (Shimadzu UV-1800).
Continuous process

Continuous process conditions were set once the conditions of the batch process were established. For this reason, only the dyes that presented the highest adsorption percentage in the batch process were tested in the continuous one (cationic green and anionic blue). Moreover, two types of columns were evaluated, a small column Afora 5831 ( nº 0 porous plate, glass key with 2/3 mm key passage, 10 mm internal diameter, 200 mm useful length) and a larger column Afora 5855 ( nº 0 porous plate, glass key with 2/3 mm key pass, 30 mm internal diameter and 500 mm useful length). The first column was used to perform experiments towards the determination of the evolution of the adsorbent adsorptions to the column pass volume (5 g of adsorbent, 30 ppm dye solution, 25 mL/min, recovered adsorbent was washed with ethyl alcohol) and, the second, to determine the influence of the flow rate (125, 43 and 23 mL/min for 20 g of adsorbent) together with the concentration of dye (20 g adsorbent with 30 and 60 ppm of dye) in the dye adsorption yield.

All methods were carried out in accordance with relevant guidelines and regulations

Results and discussion

Batch process

The influence of the amount of the chemically and physically treated orange and lemon peels (adsorbents) to the cationic dyes adsorption was firstly evaluated. To do this, cationic dyes were initially analysed by UV-Vis absorption spectroscopy in order to obtain the wavelengths corresponding to the peaks of maximum absorption. The resulting spectra indicated that the maximum absorption signal for each one of the cationic dyes were found at 652 nm, 488 nm, 415.5 and 613.5 nm for blue, red, yellow
and green, respectively, and for anionic were found at 607 nm, 491 nm and 399 nm for blue, red and yellow, respectively. Afterwards, adsorption experiments with orange-derived and lemon-derived adsorbents with the mentioned dyes were carried out. A clear liquid fraction was obtained after 45 min when 0.5, 0.75 and 1.00 g of orange-derived and lemon-derived adsorbents where put in contact with cationic dye, indicating a high adsorption capacity of both adsorbents (Fig. 2).

**Fig. 2** a) Orange-derived and b) lemon-derived adsorbents in different concentrations after adsorption process with cationic red dye.

Those liquid fractions or liquors where subsequently analysed by UV-Vis spectroscopy and the percentage of adsorbed dye was calculated for each amount of adsorbent used (Fig. 3).

**Fig. 3** Percentages of adsorbed cationic dyes according to the amounts of (left) orange-derived and (right) lemon-derived adsorbents.

Similar adsorption values were obtained for orange-derived and lemon-derived adsorbents, however the overall percentage was slightly higher in the case of orange-derived adsorbent. Blue, yellow and green cationic dyes where increasingly adsorbed with the concentration of both adsorbents, although a discreet increase of a 0.25% between 0.75 and 1.00 g of orange-derived adsorbent was spotted for the green. Concretely, in the case of yellow, differences in the adsorption were found in the nature of the adsorbent, being higher in the case of orange-derived. Contrastingly, red cationic dye was similarly adsorbed for both adsorbents, presumably indicating an important correlation between the chemical nature of the dyes to the adsorption results.
Furthermore, the capacity of the orange-derived and lemon-derived adsorbents was extensively evaluated with anionic dyes (Fig. 4).

**Fig. 4** Percentages of adsorbed anionic dyes according to the amounts of (left) orange-derived and (right) lemon-derived adsorbents.

In contrast to the results obtained for cationic, blue anionic dye was highly retained by the lemon-derived adsorbent rather the orange-derived, presenting an averaged difference of the 31.66%. Nevertheless, the concentration of adsorbent was essentially an independent parameter when values were compared within the same adsorbent for this dye. On the other hand, the percentage of red anionic dye adsorbed increased with the amount of adsorbent, similarly to yellow anionic dye. However, yellow anionic dye was the only dye that showed a higher retention with orange-derived adsorbent than the other one. Those observed differences in the adsorption with the different dyes may have probably been influenced by the stereochemistry of the molecule of the dye due to the position and the steric hindrance of the reactive groups in each case. Moreover, an amount of 1.00 g of adsorbent was established to be the more suitable concentration on dye adsorption and so, subsequent experiments were set with this parameter.

The influence of the dye concentration in the adsorption capacity of the adsorbents was also evaluated maintaining at 1.00g the amount of the adsorbent. The assay was performed with the dyes that showed the strongest and the weakest adsorption affinity to the adsorbents. Thus, green and yellow cationic dyes and blue and yellow anionic dyes were tested with different initial concentrations, ranging from 30 to 240 ppm (Table 1).
Table 1 Percentages of adsorbed dyes according to its initial concentration for orange-derived and lemon-derived adsorbents.

Both, orange-derived and lemon-derived adsorbents did not exhibit significant differences in the adsorption of green cationic dye, resulting in high adsorption efficiencies even at the higher concentrations. On the contrary, yellow cationic dye experienced the maximum retention between 90 and 120 ppm for both adsorbents, and then a moderate decay at higher dye concentrations. Due to the poorer affinity between yellow cationic dye and the adsorbents respect to the green homologue, a major quantity of the yellow was necessary to achieve similar adsorption results as green, in concordance with previous observations. In the case of anionic dyes, orange-derived adsorbent experienced a maximum adsorption peak at 90 ppm of blue dye, however at higher concentrations a progressive decreasing of this adsorption was spotted. On the other hand, when lemon-derived adsorbent was put in contact with blue anionic dye, an almost lineal decrease between 30 and 120 ppm could be observed \((y = -0.3003x+102.2, R^2 =0.97)\), indicating the highest adsorption efficiency at the lowest dye concentration. As expected due to the previous results, yellow anionic dye showed discreet adsorption results for both types of adsorbent, presenting a maximum at 60 ppm. Even though, the following assays were performed with a concentration of 30 ppm of dye.

Another parameter that may greatly influence the capacity of dye adsorption of adsorbents is the pH value. For this reason, adsorption experiments were carried out,
comparing different pH values: as-it-is, acid (pH = 4 and 2) and alkaline (pH = 10 and 12) (Fig. 5).

**Fig. 5** Percentages of adsorbed (top) cationic and (bottom) anionic dyes according to the different pH values for (left) orange-derived and (right) lemon-derived adsorbents.

Experiments with cationic dyes revealed that the capacity to adsorb red and green dyes was not significantly affected by the pH. However, blue and yellow dyes were greatly retained with an unmodified pH value, as-it-is, by both adsorbents. Orange-derived adsorbent resulted in higher values of adsorption when the nature of adsorbent was compared. Furthermore, in the case of anionic dyes, the adsorption capacity experienced a progressive decrease with the increase of the pH values for red and yellow dyes. On the other hand, the percentage of adsorbed blue dye kept almost invariable along the different values of pH. Contrary to the cationic, anionic dyes were better adsorbed by the lemon-derived adsorbent. Because in the most cases higher absorption results were obtained by the pH as-it-is, the two-type adsorbents with unmodified pH were concluded to be the most suitable for dye adsorption.

Moreover, the adsorption time or the contact time between the adsorbent and the dye solution was also determined. Adsorption times were different according to the nature of the dyes because previous results demonstrated a faster adsorption of anionic dyes than cationic. Both, orange-derived and lemon-derived adsorbents could retain an initial increasing percentage of cationic dyes within the first 30 min and then, a plateau appeared (Fig. 6). Similar behaviour was observed in the case of anionic dyes, however the plateau appeared after 15 min of contact time.
Within cationic dyes, green was the fastest and the highest percentage dye that was adsorbed by both adsorbents and yellow the lesser in all senses, in concordance with previous observed results. On the other hand, blue and yellow anionic dyes experienced a similar behaviour as the adsorption affinity assays exposed above, being the more and faster adsorbed the blue, and the lesser and slowest the yellow. Those differences were presumably related with the chemical structure and steric hindrance of the reactive groups of the dyes. Contact times of 30 min for cationic dyes and 15 min for anionic dyes were established to be suitable to obtain adequate adsorptivities with those adsorbents.

All experiments were carried out using dried orange-derived and lemon-derived adsorbents. Nevertheless, the influence of the hydration degree of the adsorbents to the capacity of adsorption was determined. To do this, same experiments performed for the determination of the adequate contact time between adsorbent and dyes were repeated with hydrated adsorbents. With the objective to achieve a ‘hydrated’ state, adsorbents were left in contact with distilled water 24 h before conducting the assay. Results demonstrated that hydrated adsorbents could retain more effectively cationic and anionic dyes, especially in the case of yellow dyes, whose affinities to the dry adsorbents were the weakest (Fig. 7). This observation could be related with the capacity to form hydrogen bonds between water and surrounding dye molecules. Optimal contact times and its behaviour appeared to be the same for hydrated than for dry adsorbents. Consequently, hydrated adsorbent was concluded to be the most suitable state to the adsorption of cationic and anionic dyes.
Fig. 7 Percentages of adsorbed (top) cationic and (bottom) anionic dyes according to the different contact times for (left) hydrated orange-derived and (right) hydrated lemon-derived adsorbents.

Continuous process

The determination of the adsorption capacities of orange-derived and lemon-derived adsorbents in a continuous process were developed after the optimum conditions were established in a batch process. Continuous process experiments were conducted using the dyestuffs that presented the highest adsorption percentages in the batch process and so, green as cationic dye and blue as anionic were tested. First, an experiment to determine the evolution of adsorption of the adsorbents according to the volume that passes through was conducted in a small column. The eluted fluid from the column was collected and used in a second elution through the recovered adsorbent, simulating the effect of a second column in series. This process was repeated a third time, simulating a third column in series. The capacity of adsorption of the adsorbents decreased as the elutions of dyestuffs developed, as expected, indicating a decrease of the adsorption yield (Fig. 8). In the case of green cationic dye, after the first elution or column, the percentage of adsorbed dye was reduced down to an 18% when orange-derived adsorbent was used and until a 2% with the lemon-derived. However, this percentage of dye adsorbed increased up to 76% and to 19% for orange-derived and lemon-derived, respectively, when the adsorbent was recovered and passed through a second serial column. At the end of the third elution, the amount of dye adsorbed raised up to 90% in the case of the orange-derived adsorbent and up to 59% in the case of the lemon-derived homologue. A preferentially adsorption of the cationic dyes by orange-derived adsorbent could be concluded.
Fig. 8 Percentages of adsorbed of (top) green cationic dye and (bottom) blue anionic dye vs. the volume of elution for (left) orange-derived and (right) lemon-derived adsorbents.

On the other hand, a 20% of blue anionic dye could be adsorbed by the orange-derived adsorbent, similarly to the lemon-derived, which was 18% (Fig. 8). Afterwards, the second column was able to retain up to 48% and 47% of dye when orange-derived and lemon-derived were used as adsorbents, respectively. Finally, after the third elution, the capacity of the orange-derived and lemon-derived adsorbents increased up to 67% and 71%, respectively. In contrast to cationic dyes, anionic dyes did not show significant contrast in adsorptivity values when the different types of adsorbent were compared.

Additionally, the evolution of the adsorption capacity of the adsorbents was investigated at different flow rates. This part of the study was performed with green cationic dye and orange-derived adsorbent due to the promising obtained results. Similarly to the previous part, three serial columns disposition was simulated, so the fluid recovered at the end of the first column pass was sequentially passed again two more times. Although the maximum flow rate studied presented an adsorption of 90%, the elution was not stable (Table 2). The stabilisation of the elution was achieved at 23 mL/min, where a 100% of the dye was adsorbed at the 2nd column. Starting from the minimum flow rate, this was progressively increased until a decreasing of the adsorption was not observed together with a stable elution. In this way, 43 mL/min was found to be the more balanced flow rate.

Table 2 Percentages of adsorbed dye according to the flow rate and column pass.
Once the flow rate was established, the adsorption capacity of the adsorbents was evaluated at different green cationic and blue cationic dyes concentration in order to observe differences in the adsorption yield. Results showed that, excepting for the elution of the green dye with lemon-derived adsorbent, more promising results were obtained when the concentration of the dye was 60 ppm (Table 3). Thus, the concentration of dye with the utmost efficiency of the adsorbent could be established. Moreover, higher adsorption percentages were obtained with orange-derived adsorbent when the nature of adsorbents was compared within the same cationic dye. Contrastingly, the most suitable type of adsorbent to retain anionic dye was the lemon-derived, in concordance with the previous observed results.

Table 3 Percentages of adsorbed dye according to the initial dye concentration for each type of adsorbent and column pass.

Conclusions

In this manuscript, the successful use of orange-derived and lemon-derived peels as adsorbents after some treatments to the adsorption of cationic and anionic dyes from textile dyeing wastewater has been demonstrated. The study has been conducted simulating two industrial processes, a batch and a continuous. From the batch process, a concentration of dye of 30 ppm, without the modification of the dyestuff, contact times between the dye and adsorbent of 30 min for cationic dyes and 15 min for anionic together with a hydrated adsorbent were the most favourable conditions. Moreover, the behaviour of orange-derived and lemon-derived adsorbents for cationic dyes has converged at concentrations of dye above 30 ppm.
In the case of the continuous process, the most suitable conditions of flow rate have been found at 43 mL/min, being the orange-derived adsorbent the more promising for cationic dyes and lemon-derived for anionic dyes. Furthermore, it has been demonstrated that the column efficiency increased with the dyed wastewater concentration, exhibiting an increase of the double of the adsorption of dye when the initial concentration increased from 30 ppm to 60 ppm for cationic dyes and more than four times with the yellow anionic dye. Finally, the recovery of the adsorbed dye by ethyl alcohol, allowing the regeneration of the column until seven times at least has been also demonstrated.

This study is a contribution to the circular economy since it takes advantage of agricultural wastes from the industry of juice production to transform them into bio-derived adsorbents towards the purification of coloured wastewaters from dyeing textile materials.

Disclosure statement

No potential conflict of interest was reported by the authors.

References


*Materials Research Express*, available at:


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Orange-derived and lemon-derived adsorbents with controlled grain for an efficient elimination of some cationic and anionic dyes

**Table 1** Percentages of adsorbed dyes according to its initial concentration for orange-derived and lemon-derived adsorbents.

<table>
<thead>
<tr>
<th>Type of dye</th>
<th>Dye</th>
<th>Adsorbent</th>
<th>Initial concentration (ppm)</th>
</tr>
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<tbody>
<tr>
<td></td>
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<td></td>
<td>30</td>
</tr>
<tr>
<td>Cationic</td>
<td>Green</td>
<td>Orange</td>
<td>96.30</td>
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<tr>
<td></td>
<td></td>
<td>Lemon</td>
<td>96.30</td>
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<td></td>
<td>Yellow</td>
<td>Orange</td>
<td>72.65</td>
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<td></td>
<td></td>
<td>Lemon</td>
<td>60.75</td>
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<tr>
<td>Anionic</td>
<td>Blue</td>
<td>Orange</td>
<td>71.5</td>
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<tr>
<td></td>
<td></td>
<td>Lemon</td>
<td>95.0</td>
</tr>
<tr>
<td></td>
<td>Yellow</td>
<td>Orange</td>
<td>48.0</td>
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<td></td>
<td></td>
<td>Lemon</td>
<td>40.4</td>
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**Table 2** Percentages of adsorbed dye according to the flow rate and column pass.

<table>
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<th>23</th>
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<tr>
<td></td>
<td>1st column</td>
<td>2nd column</td>
<td>3rd column</td>
</tr>
<tr>
<td>Adsorbed dye (%)</td>
<td>86.1</td>
<td>3.6</td>
<td>1.0</td>
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**Table 3** Percentages of adsorbed dyes according to the initial dye concentration for each type of adsorbent and column pass.

<table>
<thead>
<tr>
<th>Dye concentration (ppm)</th>
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<th>60</th>
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</thead>
<tbody>
<tr>
<td>Adsorbed dye (%)</td>
<td>Dye</td>
<td>Adsorbent type</td>
</tr>
<tr>
<td>------------------</td>
<td>--------</td>
<td>----------------</td>
</tr>
<tr>
<td>Green</td>
<td>Orange</td>
<td>98.0</td>
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<tr>
<td></td>
<td>Lemon</td>
<td>90.7</td>
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<tr>
<td>Blue</td>
<td>Orange</td>
<td>57.0</td>
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<tr>
<td></td>
<td>Lemon</td>
<td>42.1</td>
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