



An interesting environmental friendly cleanup: The excellent potential of olive pomace for disperse blue adsorption/desorption from wastewater



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ARTICLE INFO

Article history:

Received 19 July 2016

Accepted 28 January 2017

Available online 1 February 2017

Keywords:

Textile dye

Olive pomace

Wastewater treatment

Direct blue

Dye removal

Dye recover

ABSTRACT

The removal of Disperse Blue 73 from aqueous solutions, using olive pomace as adsorbent material, was investigated in a batch system with respect to contact time, pomace dosage, pH and temperature. SEM, FTIR-ATR, TG and XPS analyses appeared as powerful tools to characterize olive pomace, before and after the adsorption of dye, while UV–Visible analyses were used to quantify the amount of loaded dye on adsorbent material. The pseudo-second order kinetic model well fitted the experimental data and described the kinetic adsorption process. The dye desorption in glacial acetic acid was also obtained with the dye recovery enabling the recycle both of adsorbent material and dye itself. Five consecutive cycles of adsorption and desorption were performed and the absence of any degradation process affecting the dye after the adsorption/desorption cycles was observed. The recorded absorption spectrum, in acetic acid solution, before and after the desorption, confirmed such result. An environmentally friendly and a low cost material is thus presented, showing the excellent olive pomace potential both in disperse blue adsorption (with an efficiency of 100%) and desorption (with a mean value of 80% for each cycle). Additionally, an alternative environmental friendly use of olive oil solid residues is presented.

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1. Introduction

Over the last few years, the use of dyes, employed for several industrial processes such as paper and pulp manufacturing, plastics, dyeing of cloth, leather treatment and printing has considerably increased. Thus, due to the release of toxic textile dyes in the environment, the life of some organisms is affected with the incidence for aquatic life systems [1]. Moreover, as suggested by Malik [2] and Namasivayam et al. [3], the by-products arisen from dye reactions are highly carcinogenic and mutagenic. Indeed, the importance of the problem appears more evident considering, in accordance with the Color Index (managed by the Society of Dyes and Colorists and the American Association of Textile Chemists and Colorists), that more than 10,000 types of dyes are available in the world with a mean annual production of over 700,000 tons of dyes [4].

Generally, dyes can be classified as anionic (direct, acid and reactive dyes), cationic (basic dyes) and non-ionic (disperse dyes) [5]. In particular, besides azo-dyes, the anthraquinone dyes, belonging to the disperse ones, represent the second largest class of textile pigments occurring resistant against degradation due to their aromatic structures. Their synthesis is complex and the product costs are higher if compared with azo dyes [6]. Moreover, the degradation efficiency of anthraquinone dyes depends mainly on the microbial capacity to remove chromogenic groups [7]. Thus, it appears clear that scientific communities have the responsibility to contribute to the wastewater treatment with the aim to develop efficient dye removal techniques [5]. Recently, we have shown the excellent ability of cyclodextrins in dyes removal from wastewater [8]. More specifically, anionic azo-dyes (direct) were studied. The use of chitosan films [9] in dye removal from wastewater was also extensively studied, showing an innovative procedures to modify chitosan adsorbent improving the anionic azo-dyes uploading. However, in the point of view of a circular green economy, one of the main objectives is also the reuse of both the adsorbent materials and dyes. In fact, the latter purpose represents another important topic of research beside the "simple" removal of dyes

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from wastewater with the possibility to perform many cycles of dye adsorption/desorption, reducing at least the cost as a whole. Excellent results were obtained, for anionic dyes, considering, for example, the use of cyclodextrins.

In this paper, the attention was focused on the anthraquinone dye, Disperse Blue 73 (DB), as an example of disperse dye. An innovative material, the olive pomace (OP), as a physical adsorbent was used enabling the possibility to recover the disperse dye for several textile dyeing processes. Indeed, among treatment technologies, the adsorption has rapidly gaining importance as a method for aqueous effluents treatment. Not surprisingly, some of the advantages of adsorption process are the promising low cost regeneration of the adsorbent, the availability of known process equipment, simple operations and recovery of the adsorbed compounds [10]. Further, the agricultural waste materials have little or no economic value and often induce a discarding problem. Therefore, the use of agricultural wastes can be considered of great significance for our purpose [5]. With this regard, The European Union is the main producer and consumer of olive oil in the world. Indeed, recently, in the 2014, Bhatnagar et al. [11] reported that the global olive oil production for 2010 was estimated to be 2,881,500 metric tons. Moreover, the worldwide consumption of olive oil is increased of 78% between 1990 and 2010. Valta et al. [12] (2015) reported an increase of this production reaching the value of 3.3 million tons for the 2011/2012. The increase in olive oil production implies a proportional increase in olive mill wastes [11] and the main world producer countries are reported to be Spain, Italy, Greece, Turkey, Syria and Tunisia [13]. Indeed, olive production across the Mediterranean area has a long and prestigious past and nowadays the olive oil industries are very important in Mediterranean countries, both in terms of wealth and tradition [14]. Consequently, this area is affected by olive mill wastes pollution and many efforts are attempted to overpass this problem with innovative technologies [15]. As a consequence of such increasing trend, olive solid residues are facing severe environmental problems due to lack of feasible and/or cost-effective solutions to olive waste management. For this reason, among biomaterials, solid residues of the olive oil production could constitute a promising low-cost adsorbents [16] overcoming two problems: the elimination of olive solid residues from environment, in order to use the latter to reduce another important class of pollutants, i.e. dyes from wastewater. Not surprisingly, some of these olive oil wastes were tested as low cost biosorbents materials for several pollutant disorders [17]. Olive pomace consists of cellulose, lignin, amino acid, protein, uronic acids and polyphenolic compounds and it is generally swallowed by means of controlled spreading on agricultural soil and only a small amount of this residue is used as natural fertilizer, or as source of heat energy or as additive in animal food [18]. One of the interesting use of such olive solid wastes is to remove mainly heavy metals from water. For example Lead, Chromium, Cadmium, Zinc, Nickel, Copper, Arsenic Mercury and Iron are reported as metals extensively studied in literature [11]. Starting from 1998, the potential use of processed solid residues of olive mill products to treat drinking water containing several heavy metals in trace concentrations was explored [19]. Successively, the studies about the possible use of olive mill solid wastes as a low cost remediating adsorbent for heavy metals biosorbents, raised [16,20–24]. However, although a thorough literature survey indicates that pomace waste, or their derivatives, were used as adsorbent for removing heavy metals, its use as dye adsorbent from wastewater is limited. Indeed, to our knowledge, olive pomace was only used, from 2000 to nowadays, to prepare active carbon as adsorbent to remove different dyes [25–34]. With regard the use of olive pomace without further modifications, Banat et al. [35] presented the removal of Methylene Blue dye from aqueous solutions by olive

pomace showing that the dye uptake increased with the increase of OP amount reaching the equilibrium within 7 h. Akar et al. [18] reported dye biosorption potential of untreated olive pomace showing interesting results especially at acid pH values. Clearly in the olive pomace-related literature a gap is present when the material, as it is, without further treatments, is used for dye removal from wastewater. The presence of this gap is further confirmed by Anastopoulos et al. [36] (2015) which describe the use of residues and by-products of the olive-oil production chain for the removal of pollutants from environmental media, showing as the olive pomace is extensively used for heavy metal removal with a reduced number of manuscripts related to dyes adsorption. Starting from these considerations, in this study, olive pomace has been chosen as adsorbent material for removal and recovery, for the first time, of a disperse dye from wastewater exhibiting excellent performance with a very fast adsorption and desorption. DB is chosen as a model dye testing, in accordance with several publications [37–39], the uptake ability of olive pomace in the same conditions of industrial textiles dyeing processes: i.e. from aqueous/acetic acid DB solutions, pH 3.5, as suggested by Colorprint Fashion, a Spanish textile industry.

2. Material and methods

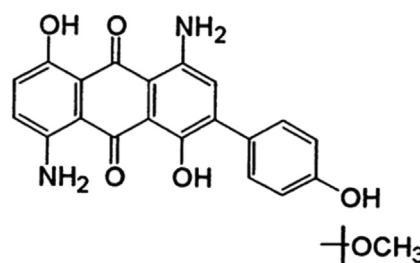
All the chemicals used were of analytical grade and samples were prepared using double distilled water. Acetic acid (99,9%), NaOH and HCl was purchased from Sigma-Aldrich. Disperse Blue 73 was received by Colorprint Fashion, S.L within the LIFE + European Project named “DYES4EVER” (Demonstration of Cyclodextrin Techniques in Treatment of Waste Water in Textile Industry to Recover and Reuse Textile Dyes) and used without further purification. DB stock solution with a concentration of 1×10^{-4} M was prepared in double distilled water containing 500 μ L of acetic acid (from a 80% stock solution) per liter of solution for mimicking the textiles dyeing conditions (proposed by Colorprint Fashion). Dilutions in double distilled water were obtained. When necessary, the pH of the various aqueous solutions was adjusted using concentrated HCl (0.5 M) and NaOH (1 M) solutions.

2.1. Disperse blue information

Color Index Number: 73; chemical formula: $C_{20}H_{14}N_2O_5$; MW: 362.34 g mol⁻¹ (see Scheme 1).

2.2. Biosorbent preparation

Biosorbent material was the solid waste of olive oil production, provided from local oil industry, Bari, Italy. The sample was repeatedly washed with deionized water, at 100 °C (until clean water was obtained), in order to remove adhering dirt and soluble impurities and, then, dried at 100 °C in an oven until constant weight. The sample was used both as obtained after the treatment



Scheme 1. Chemical structure of disperse blue 73.

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