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Optimization of an ecofriendly dyeing process using the wastewater of the olive oil industry as natural dyes for acrylic fibres

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ABSTRACT

The olive oil industry releases considerable amounts of wastewater which contains huge reserves of natural dyes. Such wastewater could successfully be used for the dyeing of acrylic fibers. The influence of the main dyeing conditions (material/liquor ratio, dye bath pH, dyeing duration, dyeing temperature) on the performances of this dyeing process were studied. The dyeing performances of this process were appreciated by measuring the color yield (*K/S*) and the fastness properties of the dyed samples. A 2^4 full factorial design method was employed in order to study the interactions between the selected dyeing process parameters and to evaluate the optimal dyeing conditions. The optimization of these dyeing process factors to obtain maximum color yield was carried out by incorporating effect plots, normal probability plots, interaction plots, analysis of variance (ANOVA) and Pareto charts. A regression model was formulated using Minitab software and fitted the experimental data very well. In addition, it was found that dyeing of acrylic enables to reduce the concentration of polyphenols so that it reduced the Chemical oxygen demand *COD*. Furthermore, the biodegradability ratio (COD/BOD_5) decreases but it was always superior to 3 which means that this aqueous waste still not biodegradable. It was also found that reusing the residual bath allowed to obtain a depth of shade very similar to the first dyeing and reduced considerably the environmental parameters (the concentration of polyphénols and *COD*).

1. Introduction

Increasing awareness of the environmental and health hazards associated with the synthesis, processing, and use of synthetic dyes have generated a renewed worldwide interest in natural dyes (Bulut and Akar, 2012). During the last decade, the use of natural dyes gained momentum due to the increased demand of food, pharmaceutical and cosmetic industries. Natural dyes comprise colorants that are obtained from animal or vegetable substances without any chemical processing. This group is mostly known to be eco-friendly, biodegradable and less allergenic compared to synthetic dyes (Ghouila et al., 2012). Besides, it can have a higher compatibility with the environment. For these raisons, a considerable number of research works are being undertaken around the world concerning the production and the application of natural dyes (Vankar et al., 2007; Shaukat et al., 2009).

The textile processing industry is one of the major users of dyes (Wesenberg et al., 2003). However, until now the application of the natural dyes in this field does not meet the expectations of all the consumers because their use caters mainly to niche products for special markets such as the eco-friendly textiles. Unfortunately, these remain as up-market products which target specific clientele who is fascinated by the sustainability and the green chemistry. The low availability and the high processing cost of natural coloring substances are the most important reasons for preventing these products from being more popular. The attempts to overcome these problems and reduce prices have mainly focused on the discovery of newer sources especially from by-products of farming and forestry as well as the valorization of several wastes from food and beverage industry in order to extract from them their coloring substances and using those colorants as natural dyes for textile dyeing. This idea is very interesting because the raw material is abundant, cheap and renewable (Prusty et al., 2010).

Another problem which may also limit the application of these dyes in the textile industry is the low dye exhaustion of the majority of natural colorants and the poor fastness of dyed fabrics on synthetic fibers compared to wool and others natural fibers because synthetic fibers are hydrophobic, highly crystalline, non-polar polymers as well as the absence of active chemical groups in their polymer structure (Gupta et al., 2011). These limitations

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generate several technical problems for producing natural-dyed synthetic textile fibers in order to fulfill the demands of more eco-friendly textile products (Sriumaoum et al., 2012).

Recently, a variety of projects about the possible use of natural dyes in synthetic fibers dyeing especially polyester and acrylic fibers have been performed by various researchers. Different techniques such as chemical modification to include amidoxime groups on polymer surface (Guesmi et al., 2012) and air atmospheric plasma treatment as well as ultraviolet (UV) excimer lamp irradiation (Kerkeni et al., 2012) were used to improve the dyeing performances of these synthetic textile fibers. In these papers, the authors described encouraging results with regard to color yield, shade, and fastness properties.

Olive mill wastewater (OMW) is a dark brown to black effluent produced during olive oil extraction. This effluent is characterized by a high organic load including sugars, tannins, pectin, lipids, polyphenols and polyalcohols (Efthalia et al., 2013). The annual OMW production of the Mediterranean olive-growing countries is estimated to over than 30 M m^3 (Tezcan Ün et al., 2008). This wastewater has a considerable pollution that occurs as a result of seasonal OMW production. In fact, it is one of the most crucial environmental issues in the Mediterranean area. Therefore, it constitutes a serious problem with severe negative impact on the soil and the water quality. Several techniques have been reported in literature to treat or valorize this effluent (El-Abbassi et al., 2013). However, for technical and economic reasons, supplementary studies and researches are still required to develop efficient systems which could be really applied in a large scale to resolve this ecological problem. The construction of artificial big ponds into which OMW is stored, waiting for its natural evaporation is until now the most common practice in the Mediterranean region. Unfortunately, this method, besides being very slow, causes subsequent unpleasant environmental pollution linked to generation of bad odors due to anaerobic activity (Saez et al., 1992).

In the previous part of this work (Meksi et al., 2012), it was found that OMW contains valuable resource of abundant natural coloring substances which could be successfully exploited as natural dye for textile coloration. The application of such wastewater as a source of natural dyes can help in the preservation of environment and also decrease the cost of natural dyeing.

The objective of the present work is to investigate the dyeing of acrylic fibers by natural dyes extracted from olive mill wastewater. The modeling and the optimization of some experimental dyeing conditions were investigated using full factorial design methodology in order to study the exhausting dyeing process. The effect of mordanting on dyeing of acrylic fibre by OMW natural matters was also studied. The environment impact of this dyeing on the OMW dye bath characteristics (Total polyphenols, Chemical oxygen demand *COD*, Biological oxygen demand *BOD_5 and the biodegradability ratio COD/BOD_5*) was also studied with the effect of multi-using of the residual dye bath on these parameters.

2. Experimental

2.1. Textile material

Acrylic fabric (Plain weave and weight, 200 g/m^2) was procured commercially. Before being used, it was soaped with 2 g L^{-1} of non-ionic detergent at 60 °C for 30 min, thoroughly rinsed and air dried.

2.2. Chemicals used

Alum, ferrous sulphate and stannous chloride were used as mordants without further purification.

2.3. Olive mill wastewaters (OMW) used

The OMW used in this study was obtained from an arranged evaporating pond which is located in the region of Monastir (Menzel Hareb) in Tunisia.

2.4. Preparation of dye bath from OMW

The used dye bath from OMW was prepared accordingly to the procedure described in Meksi et al. (2012).

2.5. Dyeing procedures

In a dye bath with various liquor ratio (10:1 - 60:1), acrylic fabric was dyed in a laboratory dyeing machine (Ahiba Datacolor International, USA) at different pH values (2–8) for different durations (15–115 min) and at different temperatures (50–110 °C). The dyed fabrics were then rinsed with cold water and washed then followed by soaping with 2 g L^{-1} of a nonionic soap, Cotoblanc RS (Bezema AG, Switzerland) at 60 °C. Finally the fabric samples were washed thoroughly with cold water, squeezed and dried. The pH values were recorded with Eutech Instruments pH 510 (Singapore).

In case of mordanting, the three known methods: pre-mordanting, meta-mordanting and post-mordanting were used. Mordant concentration of 3% (w/w with respect to the fabric) and a material to liquor ratio of 1:40 were used for all experiments. The acrylic fabrics were treated at 30 °C for 45 min.

2.6. Color evaluation

The spectral reflectance of the dyed samples was measured using SpectroFlash SF300 spectrophotometer with dataMaster 2.3 software (Datacolor International, USA). The color yield (*K/S*) values were calculated by Kubleka-Munk equation (Kubelka, 1948, 1954):

$$K\Big/S = \frac{(1-R)^2}{2R} - \frac{(1-R_0)^2}{2R_0}$$

where *R* is the decimal fraction of the reflectance of dyed fabric, R_0 is the decimal fraction of the reflectance of undyed fabric, *K* is the absorption coefficient and *S* is the scattering coefficient.

In case of dyeing with mordants, the shades may vary. So, the dyeing performances of the dyed samples were appreciated by measuring the *Sum*(*K/S*) which is calculated as follow:

$$Sum\left(K\Big/S\right) = \sum_{\lambda=400}^{700}\left(K\Big/S\right)_{\lambda}$$

CIELab coordinates (L^*, a^*, b^*, C^*, h^*, where L^* defines lightness; a^* denotes the red/green value; b^* the yellow/bleu value; C^* the saturation value and h^* is the hue) were calculated from the reflectance data for 10° observer and illuminant D_{65}.

To estimate the color differences between samples dyed with the residual dye bath and the first samples dyed with the aqueous extract of olive wastewater, the color difference was calculated as follow:

$$\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2}$$

2.7. Fastness testing

The dyed samples were tested according to standard methods. The specific tests were for color fastness to washing ISO 105-C06, color fastness to rubbing ISO 105-X12, color