ULTRASONIC-ASSISTED DYEING OF POPLAR VENEER

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Abstract. This study introduces poplar veneer ultrasonic-assisted dyeing. Response-surface methodology was adopted to perform optimum analysis of effects of ultrasonic dyeing on poplar veneer. Results demonstrated that ultrasound increased dye uptake under optimal conditions (210-W ultrasonic power, 57-min assisted dyeing time, and 30-kHz ultrasonic frequency using a dye concentration with mass fraction of 0.52% at 72°C). Under these optimal conditions, dye uptake can reach 42.4%. Compared with nonultrasonic dyeing technologies, ultrasonic dyeing technology for poplar veneer developed in this study increased dye uptake 11.2%.

Keywords: Poplar veneer dyeing, ultrasonic assistant, process optimization.

INTRODUCTION

With decreasing supply of precious tropical hardwoods, use of fast-growing timber species has become a research focus of the wood industry. Reconstituted decorative lumber and precious wood imitations prepared from dyeing fast-growing timber species have great potential in furniture-making and interior decoration.

Many researchers have conducted experiments on wood dyeing. Li et al (2009) studied dyeing of sliced veneer of plantation wood. Dedic and Zlatanovic (2001) adopted microwave drying technology to dye beech and fir wood. Guo and Guan (2010) used computer color matching to dye birch veneer artificially, but most wood dyeing technologies are heating processes. Ultrasonic dyeing technology has been adopted in the leather and textile industries as an advanced manufacturing technique to increase process efficiency, decrease process time, and improve product quality (Ahmed and El-Shishtawy 2010; Parvinzadeh et al 2010). Its special mechanical effect, thermal effect, and acoustic cavitation decrease partical size of dye as well as increase its solubility (Keun and Jae 2001), and dye uptake connects with its power, frequency, and assisted dyeing time. The most important mechanical effects of ultrasonic treatment are microjet and microstream (Luque de Castro and Priego-Capote 2007), which can make dye solution more homogeneous. Improvement in dye uptake is generally attributed to the formation of acoustic cavitation, which is gas-filled

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microbubbles or cavities in a liquid medium that produce implosive collapse and increase dye diffusion rate (Adewuyi 2001; Peters 1996). However, cavitation helps to split wood fibers into fibrils, destroys links between cellulose and noncellulosic components, and improves absorbency (Chang 2008). Until now, few researchers have reported on the application of this technology in wood dyeing.

This study investigates effects of using ultrasonic dyeing treatments on dye uptake using veneer of fast-growing poplar (*Populus euramericana cv.* '1-214') and acidic dyes as raw materials. A response-surface methodology (RSM) experiment was designed to determine optimal conditions for this dyeing process.

MATERIALS AND METHODS

Materials and Equipment

Fast-growing poplar wood was collected from South China, rotary cut into 3-mm veneer, and dried to 8% MC. Dye included acid orange II, acid red GR, acid black ATT, and a penetrant (Peregal O). The main instruments were a UV-visible spectrophotometer (UV2501-PC; Shimadzu, Kyoto, Japan), a multifrequency ultrasonic generator (20-50 kHz, custom-made), a scanning electron microscope (FEI QUANTA 200; FEI, Tokyo, Japan), and a constant temperature water bath (CS502, China).

Dyeing Effect Evaluation

On the basis of a preliminary study (Sun 2010), dye solution with a mass fraction of 0.52% was prepared with a certain ratio of dye and penetrant. Poplar veneer was dipped into the dyeing solution at 72°C under different ultrasonic power, frequency, and assisted dyeing times.

Dye uptake is an important factor in assessing dyeing effect: greater dye uptake rate means higher dye use rate. Absorbance of dye solution is assessed with a spectrophotometer (Li et al 2008). Dye uptake rate is calculated with Eq 1:

$$C_t = \frac{A_0 - A_t}{A_0} \times 100\% \qquad (1)$$

where A_0 and A_1 are absorbance of dye solution before and after dyeing, respectively (Lewis and Vo 2007).

Response-Surface Methodology Experimental Design

Based on an exploratory experiment and central composite design principles (Ravikumar et al 2006; Ferreira et al 2007) and to optimize dyeing, dye uptake was set as the responsive value, whereas ultrasonic power (X_1), assisted dyeing time (X_2), and ultrasonic frequency (X_3) were set as factors. A three-factor and three-level RSM experiment was designed (Table 1).

Eq 2 is the nonlinear quadratic model used in the response-surface experiment:

$$\mathrm{C}_t = B_0 + \sum_{i=1}^{n} B_i X_i + \sum_{i=1}^{n} B_{ii} X_i^2 + \sum_{i=1,j=1}^{n} B_{ij} X_i X_j \qquad (2)$$

where B_0 is a constant term; B_i, B_{ii}, and B_{ij} are the regression coefficients; X_i and X_j are coded levels of independent variables; and X_i^2 represents quadratic terms.

Table 2 lists all design components consisting of 17 dyeing points and five replicates at the center of the design, which were used for estimating pure error sum of squares.

Table 1. Level and code of variables chosen for response-surface methodology design.

Variable	Code	Code level		
		−1	0	1
Ultrasonic power (W)	X_1	150	200	250
Dyeing time (min)	X_3	40	60	80
Ultrasonic frequency (kHz)	X_2	24	30	36

Table 2. Design of response-surface methodology and experimental results.

Number	Ultrasonic power (W)	Assisted dyeing time (min)	Ultrasonic frequency (kHz)	Dye uptake (%)
1	−1	−1	0	34.52
2	1	−1	0	43.54
3	−1	1	0	36.12
4	1	1	0	42.48
5	−1	0	−1	33.17
6	1	0	−1	39.58
7	−1	0	1	32.12
8	1	0	1	43.15
9	0	−1	−1	38.89
10	0	1	−1	38.93
11	0	−1	1	40.73
12	0	1	1	40.72
13	0	0	0	43.05
14	0	0	0	43.47
15	0	0	0	43.19
16	0	0	0	43.46
17	0	0	0	43.47

Table 3. Analysis of variance for the fitted quadratic polynomial model of dye uptake.

Source	Sum of squares	Degrees of freedom	Mean square	*F* value	*P* value
Model	240.69	9	26.74	214.90	< 0.0001
Residual	0.87	7	0.12		
Lack of fit		3	0.24	6.20	0.0551
Pure error		4	0.039		
$R^2 = 0.9964$					

RESULTS AND DISCUSSION

Fitting Mathematical Model

Regression analysis was performed to fit mathematical models to dyeing data to determine an optimal region for responses, which were studied using Design-Expert Soft (version 7.1.3; Stat-Ease, Inc., Minneapolis, MN). In the model, *P* value was an important parameter for estimating significance of each coefficient and strength of interaction between each independent variable. Significance of *F* value was directly related to number of degrees of freedom, which was shown in the *P* value column (95% confidence level) (Cai et al 2008; Irakoze et al 2010). *P* value < 0.05 indicated model terms were significant; otherwise, they were not.

Table 3 lists analysis of variance for the fitted quadratic polynomial model of dye uptake. *P* value of the model was < 0.0001 (very low), but *F* value and *P* value of the lack-of-fit were 6.20 and 0.0551 (> 0.05), respectively, which implies that the model was not significant relative to pure error, indicating that the model equation can predict dye uptake values. Meanwhile, R^2 (determination coefficient of model) was 0.9964, which is close to 1, suggesting that the experimental results can be explained by the model very well.

Significance of each coefficient is shown in Table 4. In this instance, X_1, X_3, X_1X_2, X_1X_3, ${X_1}^2$, ${X_2}^2$, and ${X_3}^2$ are the significant model terms (*P* values are < 0.05). The second-order polynomial equation can be expressed by Eq 3. The coefficients of the quadratic model are all negative, indicating that rate of dye uptake has a maximum value.

$$\begin{aligned} C_t = 43.33 + 4.1X_1 + 0.071X_2 + 0.77X_3 \\ - 0.67X_1X_2 + 1.16X_1X_3 - 0.012X_2X_3 \\ - 3.94X_1^2 - 0.68X_2^2 - 2.48X_3^2 \end{aligned} \tag{3}$$

Response-Surface Analysis

To illustrate the relationship between dye uptake and its factors, response-surface 3D plots were used (Figs 1, 2, and 3). Figure 1 shows the effect of interaction between ultrasonic power and ultrasonic frequency on dye uptake. Within a range of ultrasonic frequency, increase in ultrasonic power increased dye uptake. When ultrasonic frequency was fixed, ultrasonic power had a significant effect on dye uptake. When ultrasonic frequency was about 30 kHz, dye uptake initially increased and then reached its maximum value as ultrasonic power elevated; beyond that frequency, dye uptake decreased slowly. When ultrasonic power was constant, changes in dye uptake were similar.

In the dyeing process, dye particles (molecules or ions) form aggregates and exist in a micellar

Table 4. Significance of each coefficient.

Variables	Sum of squares	Degrees of freedom	Mean square	*F* value	*p* value
X_1	134.64	1	134.64	1082.03	< 0.0001
X_2	0.041	1	0.041	0.33	0.5857
X_3	4.73	1	4.73	37.99	0.0005
$X_1 X_2$	1.77	1	1.77	14.22	0.0070
$X_1 X_3$	5.34	1	5.34	42.88	0.0003
$X_2 X_3$	6.25E-04	1	6.25E-04	5.02 E-04	0.9455
X_1^2	51.22	1	51.22	411.60	< 0.0001
X_2^2	1.92	1	1.92	15.43	0.0057
X_3^2	33.85	1	33.85	272.00	< 0.0001

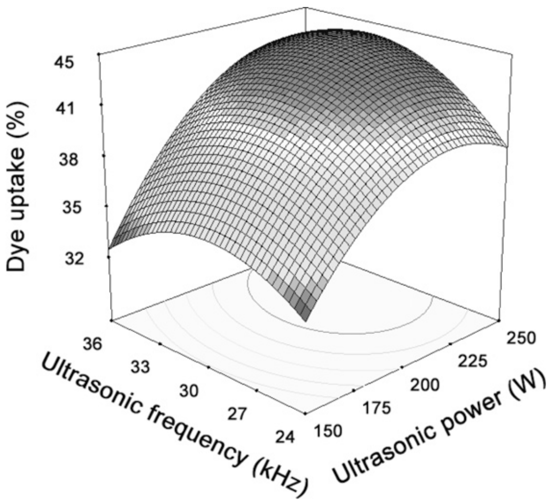

Figure 1. Response-surface plot of ultrasonic power and ultrasonic frequency and their effect on dye uptake.

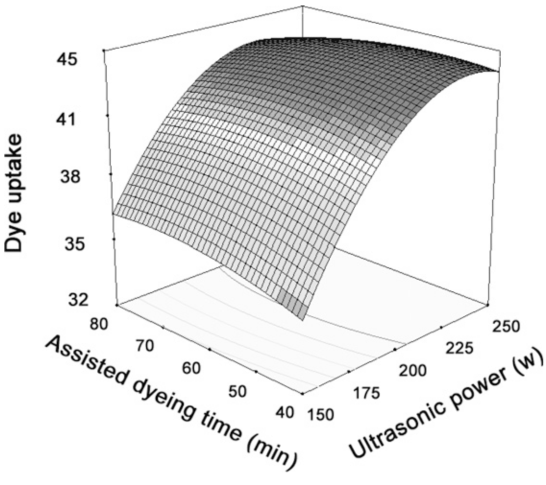

Figure 2. Response-surface plot of ultrasonic-assisted dyeing time and ultrasonic power and their effect on dye uptake.

state in the solution, which can prevent the adsorption of wood fibers on the dye particles. However, ultrasonic is accompanied by fast-moving liquid microjets, which not only depolymerizes the dye aggregates, but also crushes dye particles, thereby obtaining a highly stable dispersion solution with less than 1 μm particle size (Tsochatzidis et al 2001), which benefits absorption of wood.

Wood is a porous material; the explosive force and mechanical action of microbubble cavitation produced by ultrasonic in the catheter and pores of wood is generally attributed to stress–strain energy concentration. Acoustic cavitation formation can produce high internal pressure, which can break parenchyma cells and pit membranes and form microcracks. Figure 4 is a micrograph of poplar wood treated with ultrasonic of 200-W ultrasonic power, 30-kHz ultrasonic frequency, and a 20-min treatment time. Some microcracks (Fig 4, arrow A) appear, and many pit membranes (Fig 4, arrow B) disappear. These new cracks and pores can become new passageways for dye solution, creating many new surfaces and free radicals, which increases dye solute diffusion inside intermediate spaces of wood (Merdan et al 2004). At the same time, cavitation formation can also eliminate air in the small pores and cracks of wood, which allows dye particles to permeate into the wood interior, increasing dye absorption. Beyond a certain ultrasonic frequency, however, a higher ultrasonic power can produce more "hot spots" (Ahmed

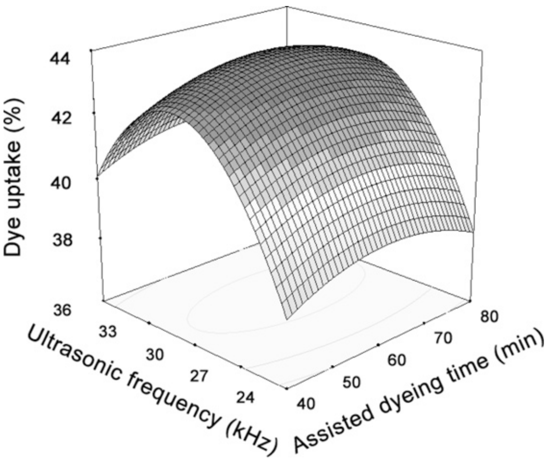

Figure 3. Response-surface plot of ultrasonic-assisted dyeing time and ultrasonic frequency and their effect on dye uptake.

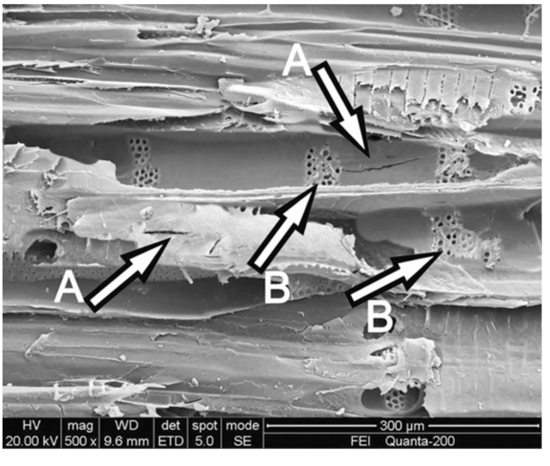

Figure 4. Micrograph of treated poplar wood (200-W ultrasonic power, 30-kHz ultrasonic frequency, and 20-min treatment time).

and El-Shishtawy 2010), accelerating oxidation of dye particles, thus decreasing dye uptake.

Figure 2 illustrates the effect of ultrasonic-assisted dyeing time and power on dye uptake. Dye uptake increased as ultrasonic-assisted dyeing time and power increased but slightly decreased with longer assisted dyeing time. Obviously, prolonged assisted dyeing time had a slight effect on dye uptake, whereas ultrasonic power had a significant effect. This was because dye particles moved with great speed for the mechanical effect and thermal effect of ultrasonic, which enhanced kinetic energy of the dye particles, thus with longer dyeing time, dye particles had greater probability of coming in contact with oxygen. The thermal effect of ultrasonic cavitation also increased oxidation of dye particles, which can lead to slightly decreased uptake. Also, dye particle absorption by wood fibers was limited because the fibers become saturated with increased dyeing time.

Figure 3 shows effects of ultrasonic frequency and assisted dyeing time on dye uptake. The effect of assisted dyeing time was similar to that in Fig 2, but the effects of ultrasonic frequency showed a great change. Under certain dyeing-time conditions, increase in ultrasonic frequency initially increased dye uptake and then decreased it rapidly. Dye uptake was greatest when assisted dyeing time was about 60 min and ultrasonic frequency was about 30 kHz.

The dye liquor had a vibration frequency similar to ultrasonic frequency with the action of ultrasound, and their vibration velocities related to their vibration frequencies. Effects of vibration, microjet, and kinetic energy of dye particles were small when ultrasonic frequency was low with some power (Yu et al 2002) and dye particle diffusion in the cell walls was limited, which led to low dye uptake. Increase in ultrasonic frequency and vibration velocity of dye particles as well as the multistage physical effects of cavitation, mechanical vibration, microjet, and acoustic streaming can increase kinetic energy of dye particles (Yang et al 2005). Of course, they can also promote deformation and damage of wood cell tissues (Fig 4), which increases permeability. However, as ultrasonic frequency further increased, the expansion-phase time of the acoustic wave became relatively short (Akalin et al 2004). This did not allow enough time for the acoustic cavitation nucleus to form and become large enough to create cavitation bubbles; hence, the effects of cavitation formation did not materialize. This means that even if cavitation bubbles formed, the compression-phase time of the acoustic wave was too short to form cavitation effects. Therefore, the effect of cavitation formation was weakened, which led to decreased dye uptake.

Process Optimization and Verification

Optimization analysis was conducted on factors that influence dye uptake using multiple regression models. Optimal technological conditions were as follows: 211.11-W ultrasonic power, 57.32-min assisted dyeing time, and 31.33-kHz ultrasonic frequency. Under these optimal conditions, dye uptake was 44.15%. A verification test was conducted three times under these conditions: 210-W ultrasonic power, 57-min assisted dyeing time, and 30-kHz ultrasonic frequency. Under these conditions, mean dye uptake was 42.4%, which was close to the predicted value, indicating that the established multiple regression model accurately predicted test results. Under similar conditions, dye uptake using nonultrasonic treatments was only 31.2%, apparently lower than that from ultrasonic dyeing treatments.

Figure 5 shows the difference between predicted dye uptake of the model and the test results. The data points were basically distributed in a straight line, which indicates that the established model had excellent predictive power.

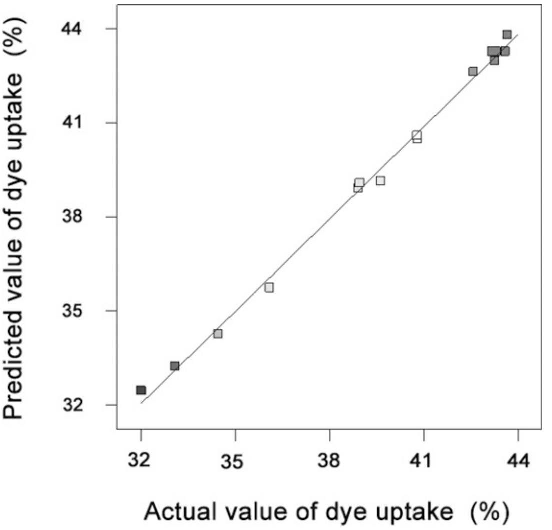

Figure 5. Discrepancy between predicted value and actual value of dye uptake.

CONCLUSIONS

Ultrasonic-assisted treatments improved poplar veneer dye uptake. Dye uptake was directly connected with ultrasonic power, frequency, and assisted dyeing time. When the mass fraction of dye liquor was 0.52% and the dye temperature was 72°C, optimal ultrasonic dyeing treatment conditions for poplar veneer were as follows: 210-W ultrasonic power, 30-kHz ultrasonic frequency, and 57-min assisted dyeing time. Under optimal conditions, dye uptake of poplar veneer was 42.4%, which is 11.2% greater than that obtained with nonultrasonic dyeing treatments under similar conditions.

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