# THE SUITABILITY OF STEAM EXPLODED *VITIS VINIFERA* AND ALKALINE LIGNIN FOR THE MANUFACTURE OF FIBERBOARD

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The main objective of this study was to explore the suitability of *Vitis vinifera* as a raw material and alkaline lignin as a natural binder for fiberboard manufacturing. In the first step, *Vitis vinifera* was steam-exploded through a thermo-mechanical vapor process in a batch reactor, and the obtained pulp was dried, ground, and pressed to produce the boards. The effects of pretreatment factors and pressing conditions on the chemical composition of the fibers and the physico-mechanical properties of binderless fiberboards were evaluated, and the conditions that optimize these properties were found. A response surface method based on a central composite design and multiple-response optimization was used. The variables studied and their respective variation ranges were: pretreatment temperature ($T_r$: 190-210ºC), pretreatment time ($t_r$: 5-10 min), pressing temperature ($T_p$: 190-210ºC), pressing pressure ($P_p$: 8-16MPa), and pressing time ($t_p$: 3-7min). The results of the optimization step show that binderless fiberboards have good water resistance and weaker mechanical properties. In the second step, fiberboards based on alkaline lignin and *Vitis vinife*ra pulp produced at the optimal conditions determined for binderless fiberboards were prepared and their physico-mechanical properties were tested. Our results show that the addition of about 15% alkaline lignin leads to the production of fiberboards that fully meet the requirements of the relevant standard specifications.

*Keywords: Vitis vinifera; Binderless fiberboards; Fibers; Steam explosion; Alkaline lignin; Mechanical properties; Binder*

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## INTRODUCTION

*Vitis vinifera* is the source of one of the most abundant agricultural waste products in Spain. It is a traditional crop cultivated for its fruit and the production of wine and covers large areas of land. After the pruning season, a large quantity of lignocellulosic material remains in the fields every year. The average pruning yield per hectare is about five tons and there are currently approximately 1,200,000 hectares of grapevines cultivated per year in Spain (Ntalos and Grigoriou 2002). Part of the pruning waste is used as fuel, but large quantities remain unused in the fields, thus increasing the risks of infestations and fire.

Moreover, the growing global population has increased the demand for various wood products and especially wood panels, leading to a continuous effort to find new resources as an alternative to forest wood. Over each of the next four to five decades, the global population is expected to increase by approximately 900 million. The per capita global consumption of wood is currently approximately 0.67 $m^3$/year (Youngquist and Hamilton 1999), and the increase in wood demand worldwide is closely tied to the increase in world population. Worldwide fiberboard production in particular has experienced rapid growth, going from 20,215,627 $m^3$ produced in 1990 to 61,861,241 $m^3$ in 2005 (FAOSTAT 2008).

The potential uses of lignocellulosic materials from agricultural waste as an alternative to wood as the raw materials in panel products have received considerable attention in the literature (Seber and Lloyd 1996; Hague et al. 1998). For example, vine shoots were studied as a possible raw material in the production of pulp for paper making (Jiménez et al. 2006). The suitability of vine pruning material for particleboard production using urea formaldehyde resin as a binder has also been explored (Ntalos and Grigoriou 2002). *Vitis vinifera* can be considered a potential lignocellulosic agricultural waste product that could replace wood as a fiber source in the production of fiberboard.

Since the 1970s, researchers have been studying various ways of using different kinds of lignocellulosic waste material for fiberboard production. Steam explosion is one of the best ways of pretreating lignocellulosic materials for use in the production of boards and composites because it preserves the fiber structure and separates the lignocellulosic material into its main components (cellulose, hemicelluloses, and lignin) (Anglès et al. 2001). In addition, lignin can easily exude from the cell wall to the fiber surface, and the fibers are well separated during steam explosion treatment (Anglès et al. 1999). The thermoplastic property of lignin is an important factor in promoting the thermal adhesion of the fiberboard pulp, from which the board is formed by means of hot pressing (Shao et al. 2009).

Due to the important role of lignin in this kind of board, several tests have already been conducted (Angles et al. 2001; Velasquez et al. 2003b, Mancera et al. 2011) to analyze the effects of replacing fiber with different kinds of technical lignins. The best results of these types of studies were obtained with kraft lignin, which is a by-product of sulfate-cooking wood chips. Kraft lignin is mainly used as a fuel and only a small amount (1–2%) made available by the pulp and paper (P&P) industry is commercially used in a wide range of products. One important application of lignin as a raw material is as an adhesive for boards in the absence of conventional phenol formaldehyde or urea formaldehyde resins. This use has been the subject of study and many patents related to it have been issued over the last few decades (Nimz 1983). For a variety of reasons these procedures have not yet led to any major applications (Pizzi 2004).

This paper explores the suitability of *Vitis vinifera* as a raw material and alkaline lignin as a natural binder for the manufacture of fiberboard. It deals with the optimization of pretreatment and pressing conditions, and examines the effects of the addition of alkaline lignin as recovered from the black liquor on the physico-mechanical properties of *Vitis vinifera* fiberboards.

## EXPERIMENTAL

### Raw Material Preparation

The *Vitis vinifera* branches came from a vineyard near Tarragona, Spain. The material was air dried and stored in jute bags. The branches were chipped to splinters smaller than 5 cm using a GA100 Black & Decker shredder. The average chemical composition of the initial raw material was reported in a previous work (Mancera et al. 2011). Alkaline lignin used in this work was derived from softwood. It was purchased from Ligno-Tech Iberica, recovered directly from the pulping liquor, and used in its powder form without further treatments. Its general features are: 3.9% moisture content, 94.1% total lignin, and 1.5% carbohydrates (El Mansouri and Salvado 2006).

### Steam Explosion

The *Vitis vinifera* chips, 150 g dry base per batch, were fed into the steam explosion reactor. The reactor is a stainless steel, cylindrical batch-type reactor with a nominal capacity of 8 L, 45 bars of pressure, and a temperature of 250ºC. The steam explosion reactor was designed by the university staff and built by Justinox. A pneumatic valve connects the reactor to a 100-liter recipient in which the pretreated material is collected after the flash expansion process. Steam is fed into the reactor at the bottom to facilitate the impregnation of the material.

The chips were then treated with saturated steam at the desired temperature (160 to 240ºC) and time (2.5 to 12.5 min.) conditions. After the set time was reached, the chips were quickly depressurized into a 100-liter recipient. The pulp obtained from this pretreatment was washed with water to rinse out the liquor derived from the pretreatment process; this liquor contains extractives and hemicelluloses. Finally, the obtained pulp was air dried at room temperature to obtain a balanced moisture content and stored in plastic for subsequent grinding and determination of its chemical composition.

### Grinding

The pretreated pulps with a moisture content of between 8 and 10% were ground and passed through a 4 mm sieve. Previous studies have shown that this procedure increases the bonding area and improves the internal bond strength (Velasquez et al. 2002).

### Board Production

The ground material was homogenized and its weight and relative humidity were measured. The material was then shaped into a forming box (150 mm long × 50 mm wide), which had previously been heated to the desired temperature, along with the press platens. The test boards were made with an objective thickness of 3 mm. After the material was placed into the mold, it was hot pressed in a three-stage cycle: (i) pressing at the desired temperature and pressure for a given period of time, (ii) a breathing period or pressure relaxation for 1 min, and (iii) pressing at the desired temperature and pressure of 12 MPa for 5 min.

The alkaline lignin-*Vitis vinifera* fiberboards were prepared following the above-reported procedure, after air drying the ground, pretreated material to 7% moisture content and mixing it with the lignin at different proportions of 5%, 10%, 15% and 20% based on the total weight of the board.

## Physical and Mechanical Characterization

The boards were characterized using European standards. The mechanical properties measured were modulus of rupture (MOR), modulus of elasticity (MOE) (EN310), and internal bond (IB) (EN319). Dimensional stability was characterized by measuring thickness swelling (TS) and water absorption (WA) (EN317). Additionally, the density of the boards was determined according to (EN323).

The boards were conditioned at 20ºC and 65% RH before any physical or mechanical test was conducted, and the dimensions of test pieces were determined based on EN325 standard (EN325). The European standards for these properties are as follows: density >800 kg/m$^3$, MOR ≥ 40 MPa, MOE ≥ 3000 MPa, IB ≥ 0.7 MPa, WA ≤ 30%, and TS ≤ 20%.

## Chemical Characterization

The original raw material and pretreated pulp were analyzed chemically to evaluate the effects of the pretreatment conditions on the composition of the fiber. Standard ASTM methods were used for this purpose and the chemical properties analyzed were moisture content (ASTM E871-82), ash content (ASTM D1102-84), and Klason lignin (ASTM D1106-96). The carbohydrates from the Klason lignin hydrolysis test were analyzed using HPLC (Yuan and Chen 1999) to determine cellulose and hemicellulose content. Acid-soluble lignin was also determined using the UV absorption method (Kaar and Brink 1991).

## Experimental Design

*Mechanical and physical properties*

The response surface method was used to study the effect of five factors on six response variables in 30 tests using a central composite design run in a single block. These factors were pretreatment temperature ($T_r$: 190-210ºC) and time ($t_r$: 5-10 min), pressing temperature ($T_p$: 190-210ºC), pressing pressure ($P_p$: 8-16 MPa), and pressing time ($t_p$: 3-7 min). The experimental factors and their levels were chosen based on previous experiences with binderless fiberboards. The response variables were the physical and mechanical properties, and the responses were analyzed using the software Statgraphics Plus 5.0.

*Chemical properties*

To study the chemical properties, the aforementioned design was used, but it was reduced to a $2^2$ central composite design, which was orthogonal and rotatable, and made up of 25 runs with 8 center repetitions.

## RESULTS AND DISCUSSION

### Optimization of Pretreatment and Pressing Conditions for Binderless Fiberboards

The results of the response surface experiment for the physico-mechanical properties and chemical composition are shown in Tables 1 and 2, respectively. For each response variable a variance analysis was performed at a confidence level of 95%. All the obtained fiberboards had densities around 1320 ± 25 Kg/m$^3$, and they were classified as High Density Fiberboard (HDF).

### Physical and Mechanical Response Variables

*Modulus of rupture and modulus of elasticity*

The modulus of rupture (MOR) and modulus of elasticity (MOE) were analyzed together because the data examined came from the same bending test. The fitted model for MOR yielded an R-squared of 0.966 and an SDR of 1.2 MPa. Only the pretreatment temperature was statistically significant for MOR at a confidence level of 95%. The modeled response surface (Fig. 1) shows that the best MOR values are obtained at high pretreatment temperatures and short pretreatment times. This can be explained by the fact that this combination can preserve the fiber structure and achieve physico-chemical modifications that enhance adhesive properties at fiber interfaces. Indeed, the lignin portion can easily be exuded from the cell wall to the fiber surface during the steam explosion treatment and undergo in situ plasticization, which contributes to the improvement of the final mechanical properties (Bouajila et al. 2005).

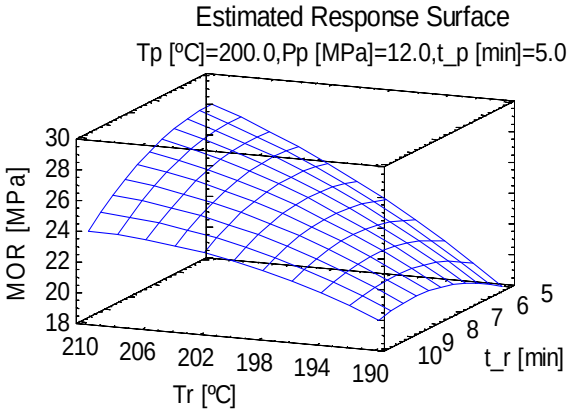


**Fig. 1.** Estimated response surface for **MOR**, $T_r$ *vs* $t_r$

The fitted model for MOE yielded an R-squared of 0.956 and an SDR of 186 MPa. In this case, two factors were statistically significant at a confidence level of 95%: pretreatment temperature and pressing pressure. The modeled response surface (Fig. 2) shows that the best MOE values were obtained at high pretreatment temperatures and short pretreatment times. The same trend was observed in the case of MOR, and therefore the same reasons and explanation given above for MOR are also valid for MOE. The modeled response surface in Fig. 3 shows that the best values of MOE are obtained at high pressing pressure and longer pressing time. This can be explained by

the improvement in the bonding strength between fibers which contributes to an increase in bending strength when fibers of the same quality are used.

**Table 1.** Results of the Physico- Mechanical Properties of Binderless Fiberboards

| | Process Factors | | | | | Response Variables | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| Run | $T_r$ (ºC) | $t_r$ (min) | $T_p$ (ºC) | $P_p$ (MPa) | $t_p$ (min) | MOE (MPa) | MOR (MPa) | IB (MPa) | WA (%) | TS (%) |
| 1 | 190 | 10 | 210 | 16 | 3 | 3439 | 19 | 0.1 | 22.2 | 19 |
| 2 | 200 | 7.5 | 200 | 12 | 5 | 3670 | 25 | 0.2 | 20.2 | 15.6 |
| 3 | 200 | 2.8 | 200 | 12 | 5 | 3410 | 22 | 0.3 | 34.7 | 18.2 |
| 4 | 210 | 5 | 210 | 16 | 3 | 3843 | 24 | 0.2 | 19.4 | 11.6 |
| 5 | 200 | 7.5 | 200 | 12 | 5 | 3615 | 24 | 0.3 | 20.8 | 16.5 |
| 6 | 200 | 7.5 | 200 | 12 | 8.7 | 3654 | 20 | 0.2 | 16.2 | 14.4 |
| 7 | 210 | 10 | 190 | 16 | 7 | 4334 | 25 | 0.2 | 16.1 | 14.9 |
| 8 | 181.4 | 7.5 | 200 | 12 | 5 | 2468 | 15 | 0.2 | 37.8 | 28.9 |
| 9 | 200 | 7.5 | 200 | 12 | 5 | 3426 | 27 | 0.3 | 20.2 | 13.4 |
| 10 | 200 | 7.5 | 218.6 | 12 | 5 | 3283 | 22 | 0.2 | 19.1 | 8.6 |
| 11 | 210 | 5 | 210 | 16 | 7 | 3778 | 23 | 0.2 | 20.2 | 10.1 |
| 12 | 200 | 7.5 | 181.4 | 12 | 5 | 3508 | 24 | 0.2 | 29.8 | 22.2 |
| 13 | 200 | 7.5 | 200 | 12 | 5 | 3733 | 23 | 0.3 | 19.2 | 15 |
| 14 | 200 | 7.5 | 200 | 12 | 5 | 3402 | 23 | 0.3 | 18.4 | 13.1 |
| 15 | 190 | 10 | 190 | 16 | 7 | 3176 | 19 | 0.2 | 27.6 | 18.9 |
| 16 | 200 | 7.5 | 200 | 12 | 5 | 3987 | 24 | 0.3 | 19.8 | 15 |
| 17 | 200 | 7.5 | 200 | 12 | 5 | 3897 | 25 | 0.3 | 18.5 | 16.6 |
| 18 | 210 | 10 | 210 | 8 | 3 | 3443 | 24 | 0.1 | 15.1 | 11.4 |
| 19 | 210 | 5 | 190 | 8 | 7 | 4487 | 26 | 0.3 | 24.9 | 8.5 |
| 20 | 200 | 7.5 | 200 | 4.6 | 5 | 3152 | 24 | 0.2 | 25.4 | 17.5 |
| 21 | 190 | 5 | 190 | 16 | 3 | 2771 | 15 | 0.2 | 47.6 | 29.2 |
| 22 | 218.6 | 7.5 | 200 | 12 | 5 | 4607 | 28 | 0.2 | 12.2 | 8 |
| 23 | 190 | 5 | 190 | 8 | 3 | 2859 | 17 | 0.2 | 48.1 | 28.3 |
| 24 | 190 | 10 | 210 | 8 | 7 | 3430 | 20 | 0.2 | 20 | 10.2 |
| 25 | 200 | 12.2 | 200 | 12 | 5 | 3040 | 20 | 0.1 | 18.9 | 10.9 |
| 26 | 200 | 7.5 | 200 | 12 | 1.3 | 3665 | 23 | 0.2 | 25.1 | 21.7 |
| 27 | 190 | 5 | 210 | 8 | 7 | 2614 | 15 | 0.2 | 24.1 | 14.6 |
| 28 | 200 | 7.5 | 200 | 19.4 | 5 | 3852 | 27 | 0.2 | 17.5 | 16.4 |
| 29 | 210 | 10 | 190 | 8 | 3 | 3245 | 27 | 0.1 | 20.4 | 16.5 |
| 30 | 200 | 7.5 | 200 | 12 | 5 | 3643 | 24 | 0.3 | 19.7 | 16 |

**Table 2.** Chemical Compositions of *Vitis vinifera* Fibers Obtained at Different Pretreatment Conditions

| | Process Factors | | Response Variables | | | | | |
|---|---|---|---|---|---|---|---|---|
| Run | $T_r$ (ºC) | $t_r$ (min) | Ash (%) | KLignin (%) | ASL (%) | Cellulose (%) | Hemicelluloses (%) | C to L Ratio |
| Original | - | - | 3.7 | 23.3 | 0.7 | 43.6 | 19.1 | 1.9 |
| 1 | 190 | 10 | 2.3 | 30.9 | 0.7 | 49.9 | 8.3 | 1.6 |
| 2 | 200 | 7.5 | 1.9 | 32.1 | 0.7 | 51.1 | 4.8 | 1.6 |
| 3 | 200 | 2.8 | 2.4 | 29.9 | 0.7 | 45 | 10.7 | 1.5 |
| 4 | 210 | 5 | 2.1 | 29.4 | 0.7 | 53.7 | 5.5 | 1.8 |
| 5 | 200 | 7.5 | 1.8 | 32.1 | 0.7 | 50.7 | 5.9 | 1.6 |
| 6 | 200 | 7.5 | 1.7 | 31.7 | 0.7 | 51.6 | 5.1 | 1.6 |
| 7 | 210 | 10 | 2 | 34.9 | 0.5 | 53.8 | 2.3 | 1.5 |
| 8 | 181.4 | 7.5 | 2.4 | 29 | 0.7 | 41.6 | 13.1 | 1.4 |
| 9 | 200 | 7.5 | 1.8 | 30.3 | 0.7 | 52 | 5.1 | 1.7 |
| 10 | 200 | 7.5 | 1.8 | 32.3 | 0.7 | 50.4 | 5.3 | 1.6 |
| 12 | 200 | 7.5 | 1.8 | 30.8 | 0.7 | 51.6 | 5.3 | 1.7 |
| 13 | 200 | 7.5 | 1.8 | 30.6 | 0.7 | 50.9 | 5.3 | 1.7 |
| 14 | 200 | 7.5 | 1.8 | 31 | 0.7 | 52.1 | 5.2 | 1.7 |
| 21 | 190 | 5 | 2.3 | 28 | 0.7 | 43.4 | 11.7 | 1.5 |
| 22 | 218.6 | 7.5 | 1.8 | 38.3 | 0.6 | 63.2 | 1.7 | 1.7 |
| 25 | 200 | 12.2 | 2.1 | 33.4 | 0.7 | 50.8 | 3.7 | 1.5 |

*Internal bond*

Internal bond (IB) is the mechanical property that accounts for the strength of the bonding between the fibers, which is an important consideration to ensure that the board will not delaminate during post-processing. The fitted model yielded an R-squared value of 0.936 and an SDR of 0.02 MPa. Only the pretreatment time was statistically significant at a confidence level of 95%. The modeled response surface (Fig.4) shows that the best IB values were obtained at short pretreatment times and high pretreatment temperatures. This can be explained by the rising quantity of fine particles that appear when the pretreatment temperature is increased (Suchsland et al. 1987); such particles augment the area available for bonding. In addition, as mentioned earlier, the lignin released in the fiber surface during the steam explosion treatment and pressing contributes to the improvement of the final mechanical properties (Bouajila et al. 2005).

*Water absorption and thickness swelling*

Water absorption (WA) and thickness swelling (TS) are physical properties related to the dimensional stability of the boards. These properties are indicative of how the boards will behave when used under severe moisture conditions. They are especially important for exterior-grade fiberboards. WA and TS were analyzed together because the data examined came from the same assay.

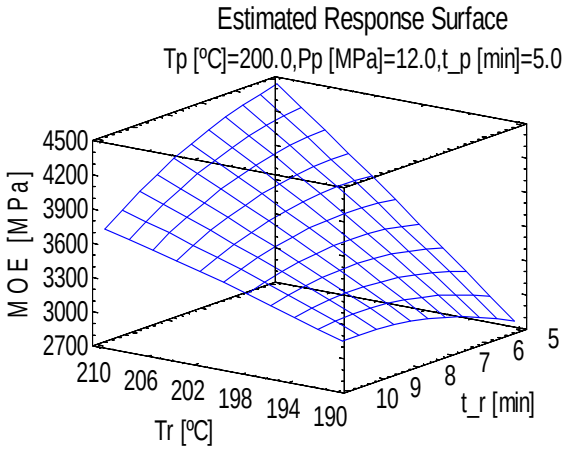


**Fig. 2.** Estimated response surface for **MOE**, $T_r$ *vs* tr

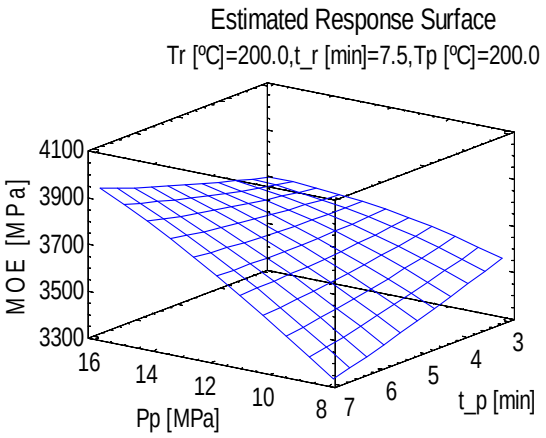


**Fig. 3.** Estimated response surface for **MOE**, $P_p$ *vs* $t_p$

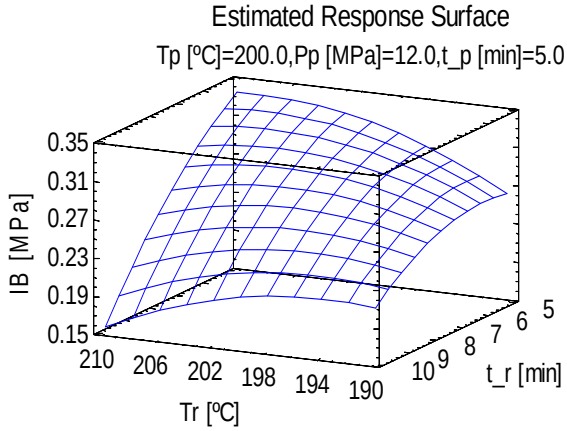


**Fig. 4.** Estimated response surface for **IB**, $T_r$ *vs* $t_r$

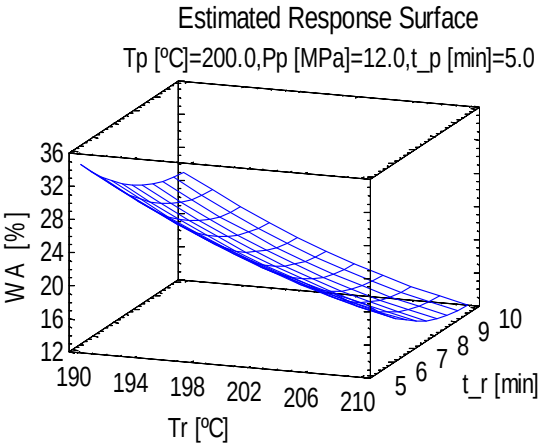


**Fig. 5.** Estimated response surface for WA, $T_r$ *vs* $t_r$

The fitted model for WA yielded an R-squared of 0.997 and an SDR of 0.84% for WA. All five factors were significant for WA. The modeled response surface in Fig. 5 shows that lower values of WA were obtained at high pretreatment temperatures and intermediate to long pretreatment times. This is because high-severity pretreatments enhance the hydrolysis of the hemicelluloses, which are largely responsible for board instability (Xu et al. 2006). Figures 6 and 7 show that lower WA values were obtained at higher pressing temperatures, higher pressing pressures, and longer pressing times. The fitted model for TS yielded an R-squared value of 0.971 and an SDR of 1.73% for TS. In this case, four factors were statistically significant: pretreatment temperature, pretreatment time, pressing temperature, and initial pressing time. The TS shows the same trend as observed for WA, and the lower values of TS were obtained at high pretreatment temperatures and intermediate to long pretreatment times.

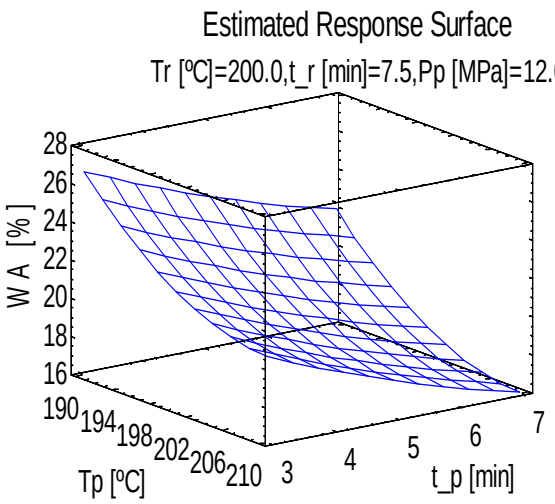


**Fig. 6.** Estimated response surface for WA, $T_p$ *vs* $t_p$

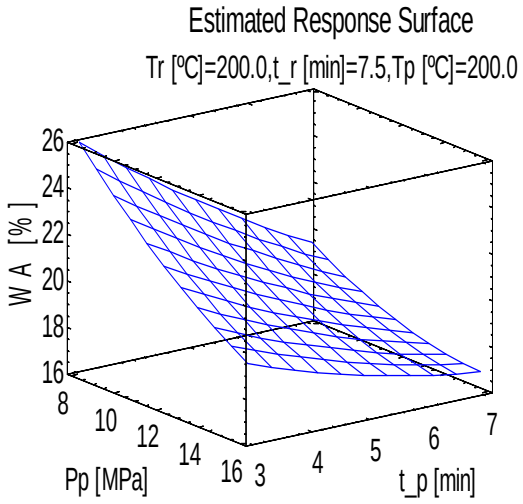


**Fig. 7.** Estimated response surface for WA, $P_p$ *vs* $t_p$

## Chemical Composition Response Variables

*Ash content*

The fitted model yielded an R-squared value of 0.846 and an SDR of 0.12%. For this variable only pretreatment temperature was found to be statistically significant. Ash accounts for mineral salts that are undesirable in the manufacture of fiberboard. Table 2 shows that the original material had a considerable amount of ash content that could negatively influence the quality of the boards. Ash content was greatly reduced by pretreatment; this reduction is due to the solubilization of the mineral salts contained in the material during the pretreatment phase. The minimum values for this response variable are found at high reaction temperatures and intermediate pretreatment times.

*Lignin, cellulose, and hemicelluloses*

Lignin, cellulose, and hemicelluloses were presented together because they came from the same hydrolysis assay. The pretreatment temperature and time have significant effects on cellulose, hemicelluloses, and lignin content. From the results presented in Table 2 it can be shown that cellulose content increased and hemicelluloses content decreased as severity intensified. The quantity of lignin increased as severity intensified. Results similar to ours with regard to the behavior of cellulose, hemicelluloses, and lignin have been obtained for other materials (Velásquez et al. 2003a, Xu et al. 2006; Mancera et al. 2008). Hemicelluloses were almost completely eliminated from the material at high pretreatment temperatures and intermediate to long pretreatment times.

## The Relationship between Chemical Composition and Physico-Mechanical Properties

It is well known that the dimensional stability of fiberboards is related to partial hemicellulose hydrolysis, because hemicelluloses are very hydrophilic. Figure 8 shows that, as expected, WA decreased as the hemicellulose content decreased. The same trend

was true for TS. Some authors (Suchsland et al. 1987; Velásquez et al. 2003a; Xu et al. 2006) have obtained similar results with other materials.

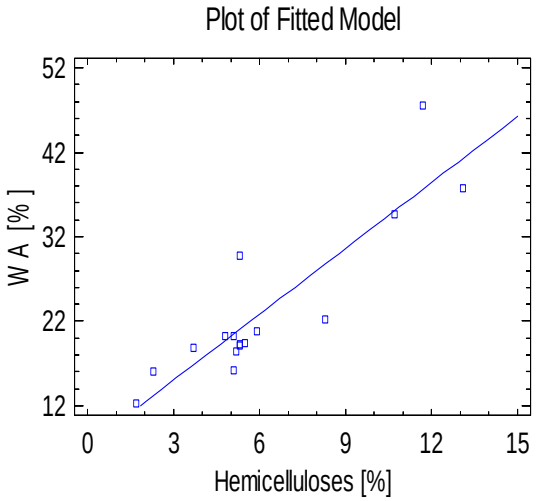


**Fig. 8.** Relationship between WA and hemicelluloses

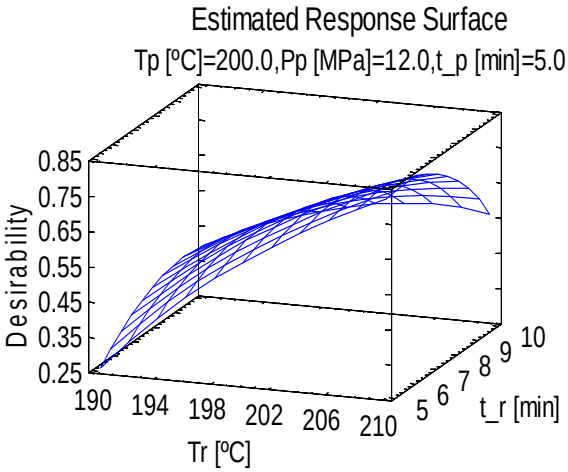


**Fig. 9.** Estimated response surface for desirability, $T_r$ *vs* $t_r$

## Multiple Response Optimizations

Multiple response optimizations determine the combination of levels for the experimental factors that simultaneously optimize several response variables. The procedure consists of building a desirability function based on the fitted models of each factor to be optimized. The goals for each of the responses were set as follows: MOR, MOE, and IB to be maximized and WA and TS to be maintained as close to zero as possible. The optimum value of desirability was 0.902 over 1 for the following factor levels: $T_r$= 218ºC, $t_r$= 6 min, $T_p$= 205ºC, $P_p$= 12 MPa, and $t_p$= 4 min. Figure 9 shows that high pretreatment temperatures and intermediate times are the best choice for simultaneously preserving the fiber structure and encouraging hemicellulose hydrolysis and the release of the lignin on the fiber surface.

A set of fiberboards were prepared using the combination of factors provided by the multiple response optimization. These fiberboards fully met European standards, except in their MOR and IB values. The mean values for the board properties were: density= 1382 kg/m$^3$, MOR= 25 MPa, MOE= 4135 MPa, IB= 0.14 MPa, WA= 12.5%, and TS= 9%. The multiple response optimization model studied suggests that the best physico-mechanical properties for the boards are found at high pretreatment temperatures and intermediate pretreatment times. The model also suggests using intermediate pressing temperatures in combination with intermediate to short pressing times and intermediate pressing pressures in the first pressing step. In the first pressing stage, moisture is vaporized and the lignin is redistributed over the fibers, where the chemical bonds are developed. After the relaxation step, in the third pressing step, the internal defects generated during relaxation are corrected and the moisture remaining on the board is vaporized.

## Effects of Alkaline Lignin as a Natural Binder on Fiberboard Properties

Table 3 shows the physico-mechanical properties for *Vitis vinifera* fiberboards produced at the previously determined optimum conditions at different lignin addition levels. The results show that there was no great difference between the densities of fiberboard produced without lignin and those produced with different percentages of lignin. As expected, increasing the amounts of alkaline lignin added had a beneficial effect on MOR, obtaining results as much as twice as high as the results obtained without lignin (25 MPa) when 20% lignin was added (50 MPa). Increasing the quantities of alkaline lignin improved MOE, similar to the behavior seen with MOR, but for MOE the best values were found at an addition of only 15% alkaline lignin. The required value for MOR was reached by adding just 5% alkaline lignin. However the IB of fiberboard produced using 5% of lignin was still far from the values required by the European standards for fiberboard. It showed significant improvement when the amount of lignin is increased and with 15% lignin replacement, *Vitis vinifera* fiberboards produced fully met the requirements of the relevant standard specifications.

Since the values of mechanical properties depend on the bonding strength among fibers and individual fiber strength, better inter-fiber bonds should contribute to an improvement in bending strength when fiber of the same quality is used. These inter-fiber bonds are due mainly to: (i) hydrogen bonding between fibers, (ii) the condensation reaction in lignin, (iii) the cross-linking reaction between lignin and polysaccharides, and (iv) the formation of covalent bonds between the constituents of lignocellulosic polymers (Back 1991; Suzuki et al. 1998; Okuda et al. 2006a, b; Zhou et al. 2011). It is well known that covalent bonds result in intermolecular forces that are much stronger than those of hydrogen bonds. In addition, fibers with lignin-rich surfaces improve the mechanical properties of binderless fiberboards through the mechanical entanglement of the melted lignin molecules under pressure and temperature, possibly accompanied by the formation of covalent bonds (Okuda et al. 2006a,b; Quintana et al. 2009).

**Table 3.** Results of Physical and Mechanical Properties of Fiberboards Prepared with Lignin Addition at Different Levels

| (%) Lignin added | Density (kg/m$^3$) | MOR (MPa) | MOE (MPa) | IB (MPa) | WA (%) | TS (%) |
|---|---|---|---|---|---|---|
| 0 | 1382 | 25 | 4135 | 0.14 | 12.5 | 9.0 |
| 5 | 1367 | 40 | 5100 | 0.30 | 11.8 | 8.8 |
| 10 | 1368 | 42 | 5554 | 0.51 | 9.2 | 7.2 |
| 15 | 1368 | 45 | 6116 | 0.70 | 9.0 | 4.0 |
| 20 | 1362 | 50 | 5628 | 1.06 | 3.6 | 1.8 |

From the results of Table 3 it ca be shown that water absorption was improved by increasing the quantities of alkaline lignin added externally. At a 20% replacement of fibers with alkaline lignin, the fiberboards achieve the lowest WA value. It is well known that lignocellulosic materials absorb water by means of hydrogen bonding between water and hydroxyl groups of cellulose, hemicelluloses, and lignin in cell walls (Rowell et al. 1976). This therefore shows that externally added lignin can reduce the degree of water absorption of *Vitis vinifera* fibers. This may be attributed to the non-polar hydrocarbon

chains and aromatic rings in lignin molecules. Lignin might reduce the absorption of water in two different ways: (i) by bulking the cell wall or (ii) by plugging the lumen of the cell wall. If the cell wall is bulked by lignin, then in conjunction with its hydrophobicity, water would not be able to enter and bulk the cell wall. The second possibility is more likely, because lignin is considered too large to enter the cell wall.

Thickness swelling of lignocellulosic material occurs when the cell wall is bulked by water. The results clearly show that fiberboards without lignin or with less incorporated lignin (5%) present high TS values. Added lignin is clearly able to reduce the extent of swelling of fiberboards, since this property is reduced as levels of lignin increase. Therefore, the results clearly indicate that lignin can reduce the amount of water going into and swelling the cell walls. The explanation is similar to that provided with regard to the water absorption results.

## CONCLUSIONS

1. It was possible to produce fiberboards using *Vitis vinifera* as a raw material and alkaline lignin as a natural binder that meet the European standards for interior-use fiberboards.
2. Both the steam explosion pretreatment and hot pressing had a great influence on the final physico-mechanical properties of the fiberboards obtained.
3. Increasing the severity of the pretreatment phase improved the physical properties (WA and TS) of the boards. Similarly, hemicelluloses and ash contents of the pretreated fibers clearly decreased as severity increased, leading to lower hygroscopicity and reducing the content of abrasive materials, which are undesirable in the fabrication of fiberboards.
4. Pretreated vines generally have higher net cellulose and lignin contents than original materials because the pretreatment phase reduces hemicellulose content.
5. The multiple response optimization models have been useful in identifying the most suitable levels of the process factors in order to manufacture the best fiberboards, particularly for the difference found between the optimization trends for physical and mechanical properties.
6. The addition of alkaline lignin in different amounts helps to improve the physical and mechanical properties of fiberboard, enough to comfortably meet the relevant international standard specifications for fiberboards.

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