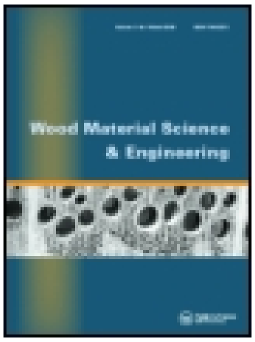

Wood Material Science & Engineering

ISSN: 1748-0272 (Print) 1748-0280 (Online) Journal homepage: https://www.tandfonline.com/loi/swoo20

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WOOD MATERIAL SCIENCE & ENGINEERING
https://doi.org/10.1080/17480272.2020.1751279

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ORIGINAL ARTICLE

Medium-density fibreboards bonded with phenol-formaldehyde resin and calcium lignosulfonate as an eco-friendly additive

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ABSTRACT
The potential of addition of calcium lignosulfonate as an eco-friendly additive with a phenol-formaldehyde (PF) resin, at different ratios, to produce medium-density fibreboards (MDF) with acceptable properties, was investigated in this work. The fibreboards were fabricated in the laboratory with low PF resin percentage (3%, 4%, 5%), while the addition levels of calcium lignosulfonate varied from 5% to 15% (on the dry fibres). The physical and mechanical properties of the fibreboards were evaluated, and further statistically analysed in order to determine the optimal values of PF resin content and lignosulfonate addition for fulfilling the European standards. It was shown that at the low PF resin content (3%), addition of lignosulfonate should not exceed 10% to avoid deterioration in the mechanical properties. It was concluded that PF resin content of 3.5% can be recommended for the production of lignosulfonate-PF bonded MDF panels to comply with the EN standard requirements.

ARTICLE HISTORY
Received 11 March 2020
Revised 24 March 2020
Accepted 31 March 2020

KEYWORDS
Medium-density fibreboard (MDF); phenol-formaldehyde resin (PF resin); calcium lignosulfonate; formaldehyde emission; lignosulfonate-PF resin; bio-adhesives

Introduction

Concerns respecting the hazardous formaldehyde emission from wood-based panels, especially in indoors, and the increased environmental awareness relating with the sustainability of raw materials and end products are the main driving forces for shifting the scientific and research interest from the conventional petroleum-derived formaldehyde-based adhesives towards new bio-based adhesives ('bio-adhesives') for the production of eco-friendly wood panels (Dunky 2004, Frihart 2005, Pizzi 2006, Kües 2007, Papadopoulou 2009, Navarrete *et al*. 2010, Carvalho *et al*. 2012, Ferdosian *et al*. 2017, Hemmilä *et al*. 2017, Valyova *et al*. 2017, Mantanis *et al*. 2018, Kumar and Pizzi 2019a, Taghiyari *et al*. 2020).

For wood-based panels, very low formaldehyde emission, e.g. below 4 mg/100 g (EN ISO 12460-5), is very difficult to achieve when using conventional adhesives based on urea, phenol and/or melamine, even if adding scavengers such as salts, bisulphites, phosphates, or other compounds with excellent capability to capture free formaldehyde (Alexandropoulos *et al*. 1998, Eom *et al*. 2006, Kim *et al*. 2006, Park *et al*. 2008, Boran *et al*. 2011, Costa *et al*. 2012, Costa *et al*. 2013a, 2013b, Funk *et al*. 2017, Kumar and Pizzi 2019a, 2019b). One of the most advanced 'green' solutions to overcome this problem is the use of formaldehyde-free bio-based adhesives, which do not significantly increase the production costs, and at the same time, result in wood-based panels with acceptable physical and mechanical properties (Pizzi 2006, El Mansouri *et al*. 2007, Nasir *et al*. 2014, Hemmilä *et al*. 2017, Nordström *et al*. 2017, Papadopoulou *et al*. 2017, Sepahvand *et al*. 2018, Hosseinpourpia *et al*. 2019, Ndiwe *et al*. 2019). Coming from the pulp industry, lignin-based byproducts, such as lignosulfonates ($R\text{-}SO_3H$), are one of the most promising eco-friendly additives for incorporation into conventional formaldehyde-based adhesive systems for wood-based panels (Geng and Li 2006, Mancera *et al*. 2011, Zouh *et al*. 2011, Ghaffar and Fan 2014, Klapiszewski *et al*. 2017, 2018, Nordström *et al*. 2017, Mantanis *et al*. 2018, Hazwan Hussin *et al*. 2019, Zhang *et al*. 2019).

The main research interest in lignin byproducts, like lignosulfonates, is high because of the phenolic structure of lignin itself. Hence, these compounds can be used as a partial replacement of phenol (C_6H_6O) in the synthesis of new lignosulfonate-PF resins, suitable for wood-based panels (Dunky 2004, Pizzi 2006, Papadopoulou 2009, Ibrahim *et al*. 2011, Ferdosian *et al*. 2017). The phenol replacement typically is far below 50%, since the addition of lignin lowers the reactivity of the resin, thus leading to increased reaction times (Danielson and Simonson 1998). Besides that, in MDF production, there are certain technical difficulties when adding the lignin-based adhesives in the fibre mass. With lignosulfonates, this problem is eliminated as they are all in the form of water-based solutions (Yotov *et al*. 2017).

It is noted that a major disadvantage for the wider industrial use of lignosulfonates in PF resins is the decreased moisture resistance of the produced fibreboards (Dumitrescu *et al*. 2009); this can be resolved by increasing the hot-press temperature and/or extending the pressing time (Antov *et al*. 2019, Savov *et al*. 2019), resulting thus in increased production costs. Consequently, it is imperative to find solutions by combining lignosulfonates with synthetic resin systems, such as

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phenol-formaldehyde or urea-formaldehyde resins (Guo *et al.* 2015, Podschun *et al.* 2016, Savov and Mihajlova 2017a, 2017b, Hemmilä *et al.* 2019).

In addition, the main disadvantages of PF resins compared with the urea-formaldehyde resins, are their high cost and the necessity to extend the pressing time (Thoemen *et al.* 2010). Despite these, PF resins provide higher board strength, significantly increased hydrophobic properties, and permanent resistance under humid climatic conditions (Dunky 2004, Pizzi 2006, Mantanis *et al.* 2018, Kumar and Pizzi 2019b). Further, phenolic resins have an excellent compatibility with lignin-originating compounds when these are used as eco-friendly additives (Cavdar *et al.* 2008, Da Silva *et al.* 2012, Papadopoulou *et al.* 2017, Solt *et al.* 2018).

Therefore, the aim of the research work was to investigate the potential to decrease the gluing content of PF resin, below the level of 8-10%, by incorporating calcium lignosulfonate into the glue composition in order to produce thin-type medium-density fibreboards with acceptable physical and mechanical properties.

Materials and methods

In the laboratory, MDF panels were fabricated at a thickness of 8 mm and target density of 850 kg/m^3. Three PF resin contents (3%, 4%, 5%) and three addition levels (5%, 10%, 15%) of calcium lignosulfonate, based on the dry weight of fibres, were applied. Calcium lignosulfonate was selected because previous studies (Savov and Mihajlova 2017a, 2017b, Antov *et al.* 2019) have shown very promising results. Industrially produced wood fibres, composed of European beech (*Fagus sylvatica* L.), common oak (*Quercus robur* L.) and white poplar (*Populus alba* L.), oven dried to 11% moisture content, were provided by Welde Bulgaria AD (Troyan, Bulgaria). The glue formulation was comprised of a PF resin (dry solids content: 50.3%), a product of Dynea (Norway), and calcium lignosulfonate additive named LignoBond DD® (Borregaard, Norway), which had the following properties (calcium content: 6%; reduced sugars: 7%; ash content: 16%; total solids content: 93%; acidic factor in 10% solution – pH = 4.3 ± 0.8; bulk density: 550 kg/m^3). The PF resin was used at 30% concentration.

Wood fibres were mixed with the adhesive in a high-speed glue blender (850 min^{-1}). Hot pressing was performed in a laboratory press (PMC ST 100, Italy). The applied press factor was 1 min/mm of panel thickness, while the press temperature was 200°C. The pressing regime applied was as follows: in the first stage, the pressure was increased to 3.0 MPa for 1 min, then it was gradually decreased to 1.2 MPa for 2 min, followed by decreasing the pressure to 0.5 MPa for 4 min. The last pressing period was carried out at the pressure of 1.4 MPa for 1 min.

The physical and mechanical properties of fibreboards were determined according to the standards EN 310, EN 317, EN 322 and EN 323 (European Committee for Standardization). Thickness swelling and water absorption tests were carried out for 24 h. A universal-type testing machine Zwick/Roell Z010 was utilised for the mechanical property tests. For each parameter, eight (8) fibreboard samples were used for testing. Variational and statistical analyses of the results were carried out by using the specialised software *QstatLab* 6.0.

Results and discussion

A summary of the physical and mechanical properties of MDF panels bonded with reduced PF resin contents and calcium lignosulfonate is presented in Table 1. The density of the fibreboards varied from 841 kg/m^3 to 853 kg/m^3, close to the targeted value. The difference in this important parameter of the MDF panels was significantly below 5%; thus, it will not influence the other physical and mechanical properties.

The regression equations respecting the effect of PF resin content and lignosulfonate addition on the thickness swelling of boards in a coded (Equation (1)) and explicit (Equation (2)) form, are as follows:

$$\hat{G}t = 15.06 - 1.30.X_1 - 1.42.X_2 - 0.55X_1.X_2 + 1.13X_1^2 - 0.25X_2^2 \ (\%) \quad (1)$$

where $\hat{G}t$ is the predicted value of the thickness swelling (24 h) (%), X_1 – PF resin content (%), X_2 – lignosulfonate addition (%)

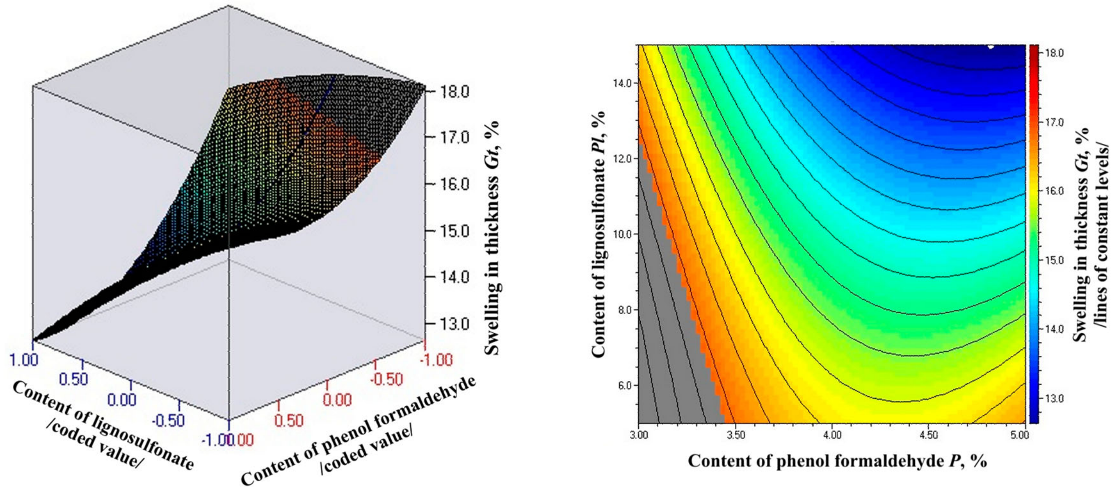
$$\hat{G}t = 35.78 - 9.24P_f + 0.35P_l - 0.11.P_f.P_l + 1.13P_f^2 - 0.01P_l^2 \ (\%) \quad (2)$$

where P_f – PF resin content (%), P_l – lignosulfonate addition (%) and coefficient of determination, $R^2 = 0.98$.

Graphical representation of the effects of PF resin percentage and lignosulfonate addition on the thickness swelling of MDF panels, in relation with Equations (1) and (2), is presented in Figure 1.

Table 1. Physical and mechanical properties of MDF panels produced in this work.

PF resin content (%)	Ligno-sulfonate addition (%)	Density ρ (kg/m^3)	Thickness swelling (24 h), *Gt* (%)	Bending strength, *fm* (N/mm^2)	Modulus of elasticity, *Em* (N/mm^2)	Water absorption (24 h), *A* (%)
3	5	845 ± 15	18.1 ± 0.4	34.1 ± 1.0	4,024 ± 190	66.3 ± 2.0
3	10	841 ± 11	17.6 ± 0.4	33.2 ± 1.0	4,202 ± 180	59.7 ± 2.7
3	15	842 ± 19	16.3 ± 0.7	28.9 ± 1.2	3,730 ± 240	57.1 ± 2.0
4	5	846 ± 15	16.4 ± 0.4	33.1 ± 1.2	4,476 ± 130	62.9 ± 1.5
4	10	852 ± 9	14.8 ± 0.7	35.1 ± 1.3	4,356 ± 180	57.7 ± 1.4
4	15	847 ± 6	13.5 ± 0.7	27.2 ± 1.1	4,276 ± 210	55.5 ± 2.8
5	5	842 ± 15	16.5 ± 0.7	35.2 ± 1.4	4,181 ± 200	63.8 ± 3.0
5	10	846 ± 8	15.1 ± 0.7	32.8 ± 1.3	4,305 ± 180	58.6 ± 2.6
5	15	838 ± 18	12.6 ± 0.6	32.4 ± 0.9	4,414 ± 130	55.8 ± 3.0
10	0	853 ± 19	12.3 ± 0.4	40.4 ± 1.6	4,894 ± 143	51.1 ± 2.3



a) b)

Figure 1. Variation of the thickness swelling of MDF panels depending upon the PF resin content and the lignosulfonate addition: (a) coded form and (b) explicit form.

Therefore, it was determined that increasing the PF resin content and the lignosulfonate addition resulted in decreased values of the MDF thickness swelling, within the studied variation range. The optimal values of the parameters were evaluated as: 4.82% PF resin content, and 15% lignosulfonate addition, i.e. close to the upper limit factor values. Nevertheless, all fibreboards produced with 4% PF resin fulfilled the standard requirements for thickness swelling of MDF (EN 622-5), for use in dry conditions (i.e. below 17%). Noticeably, Savov *et al.* (2019) have reported that MDF panels bonded with different lignosulfonate contents (20%, 30%, 40%) have also met the respective European standard requirements, for applications in dry conditions. As seen in Figure 1(b), the produced MDF panels bonded with 3% PF content can comply with the EN standards only at lignosulfonate addition level ≥12.5%.

The regression equations respecting the effect of PF resin content and the lignosulfonate addition on the bending strength (MOR) of fibreboards, in a coded (Equation (3)) and explicit (Equation (4)) form, are presented below:

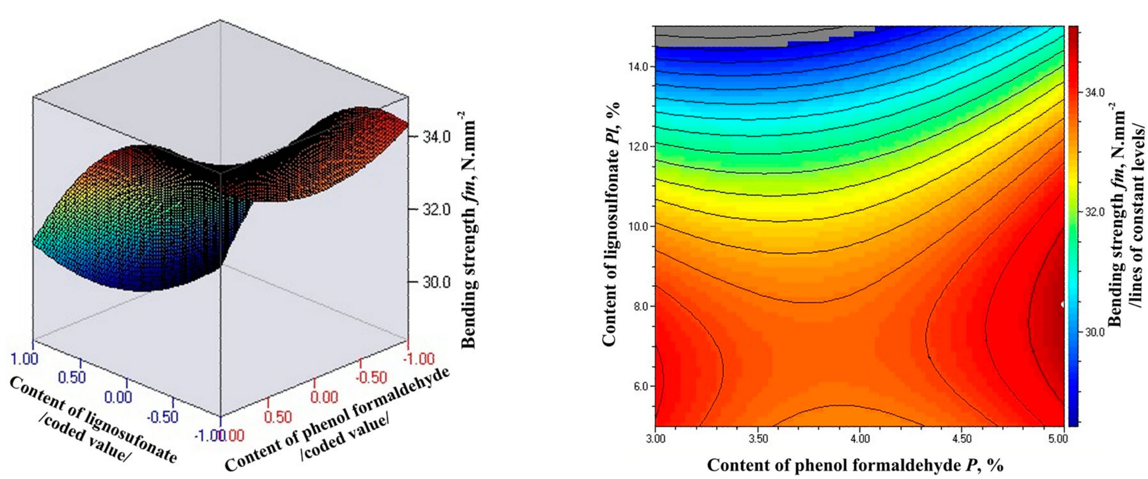
$$\hat{f}m = 33.07 + 0.71X_1 - 2.3X_2 + 0.61X_1.X_2 + 0.94X_1^2 - 1.89X_2^2 \ (\mathrm{N/mm^2}) \quad (3)$$

where $\hat{f}m$ is the predicted bending strength (MOR) value (N/mm^2), X_1 – PF resin content (%), X_2 – lignosulfonate addition (%)

$$\hat{f}m = 47.18 - 8.02.P_f + 0.56.P_l - 0.12.P_f.P_l + 0.93P_f^2 - 0.08P_l^2 \ (\mathrm{N/mm^2}) \quad (4)$$

where P_f – PF resin content (%), P_l – lignosulfonate addition (%) and coefficient of determination, $R^2 = 0.78$.

Graphical representation of the effects of PF resin content and the lignosulfonate addition on the bending strength of MDF panels, in relation with Equations (3) and (4), is presented in Figure 2.



a) b)

Figure 2. Variation of the bending strength (MOR) of MDF panels depending upon the PF resin content and the lignosulfonate addition: (a) coded form and (b) explicit form.

The MOR of the laboratory-produced MDF panels varied from 27.2 to 35.2 N/mm^2, while the respective value of the control panels (bonded with 10% PF resin) was 40.4 N/mm^2 (Table 1). Hence, the decrease in MOR varied from 12.8% to 32.6%. Due to its macromolecular nature, lignin acts as a structural backbone in the resin prepolymer (Podschun *et al*. 2016). The highest MOR values were accomplished for MDF panels produced with 5% PF resin and 5% lignosulfonate. On the other hand, the lowest MOR values were obtained for fibreboards fabricated with 3% PF resin and 15% lignosulfonate. The relative difference between the MOR values in these panels was approx. 30%. To note, the MDF panels bonded with 5% PF resin and 5% lignosulfonate exhibited MOR strength which was 14% lower, as compared with that of control panels produced only with 10% PF resin (Table 1).

It became obvious that, at all levels of PF resin content, the increase of lignosulfonate addition from 5% to 10% resulted in similar MOR strength values (Table 1), while further increase of lignosulfonate addition, from 10% to 15%, resulted in lower bending strength of the panels. This might be attributed to the increased moisture content of the pressed mat material, and higher vapour-gas mixtures at the higher lignosulfonate addition levels. This should also have had a negative effect on the hydrophobic properties of the panels, which though, was not observed. A more likely explanation may be the presence of more sugars in the lignosulfonate (7%), which turned out in increased hydrophobic properties of the MDF panels. In addition, a clear decrease in MOR was observed, when the lignosulfonate content exceeded 10%. Quite similar results have been reported by Savov and Mihajlova (2017b), when they investigated the mechanical properties of MDF bonded with 5% urea-formaldehyde (UF) resin and calcium lignosulfonate (0% to 20% addition levels).

The produced fibreboards exhibited very satisfactory bending strength values, meeting the EN 622-5 requirements for MDF – general application in dry conditions (MOR ≥ 23 N/mm^2), and also for MDF – general application in humid environment (MOR ≥ 27 N/mm^2). All fibreboards, except for those bonded with 3% and 4% PF resin and 15% lignosulfonate addition, met the standard requirements for MDF – use in load bearing applications in dry conditions (minimum MOR: 29 N/mm^2). According to recent findings of Antov *et al*. (2019), for MDF boards bonded exclusively with lignosulfonate, as binder, such a requirement can be reached at a minimum lignosulfonate addition of 26%. The maximum MOR strength recorded in this work, i.e. 35.2 N/mm^2, was realised at 5% PF resin content and 5% lignosulfonate addition.

The regression equations respecting the effect of PF resin percentage and the lignosulfonate addition on the modulus of elasticity (MOE) in bending, in a coded (Equation (5)) and explicit (Equation (6)) form, are as follows:

$$\hat{E}m = 4439 + 157.X_1 - 44.X_2 + 132.X_1.X_2 - 227.X_1^2 - 104X_2^2 \ (\text{N/mm}^2) \quad (5)$$

where $\hat{E}m$ is the predicted value of the modulus of elasticity (MOE) (N/mm^2), X_1 – PF resin content (%), X_2 – lignosulfonate addition (%)

$$\hat{E}m = 907 - 1708.P_f - 31.P_l + 26.P_f.P_l - 227P_f^2 - 4P_l^2 \ (\text{N/mm}^2) \quad (6)$$

where P_f – PF resin content (%), P_l – lignosulfonate addition (%) and coefficient of determination, $R^2 = 0.86$.

Graphical representation of the effects of PF resin content and the lignosulfonate addition on the modulus of elasticity (MOE) of MDF panels produced, according to Equations (5) and (6), is presented analytically in Figure 3.

The MOE of the produced MDF panels had very high values, ranging from 4476 to 3730 N/mm^2. These values are significantly beyond the European requirements (EN 310) for MDF – use in load bearing applications in humid environment (≥3000 N/mm^2). Quite comparable results, i.e. MOE values ranging from 2868 to 4254 N/mm^2, have been reported by Savov and Mihajlova (2017b) in their work on

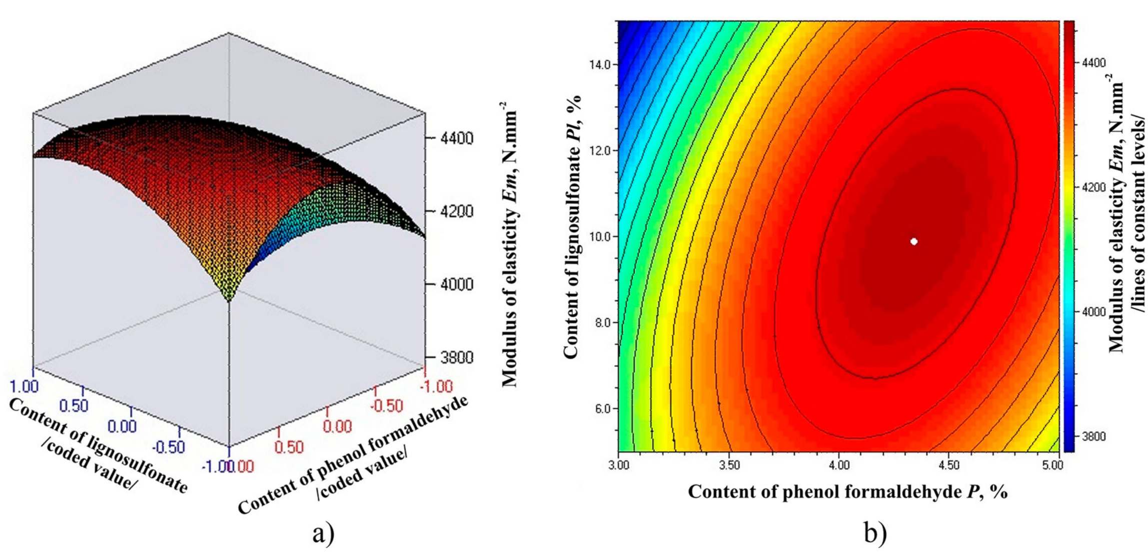


Figure 3. Variation of the modulus of elasticity (MOE) of MDF panels depending on the PF resin content and the lignosulfonate addition: (a) coded form and (b) explicit form.

eco-friendly MDF panels bonded with 5% UF resin content, and 0% to 20% calcium lignosulfonate.

For MDF panels produced with 4% and 5% PF resin content, the modulus of elasticity was not significantly affected by the increase in the lignosulfonate content. A rather decreasing trend in MOE was observed in the fibreboards fabricated with 3% and 4% PF resin as the lignosulfonate percentage increased from 10% to 15%. Therefore, it seemed that the MOE property was less dependent upon the lignosulfonate addition (%), in the glue mixture.

According to the statistical analysis, the optimal value of 4476 N/mm^2 for this indicator (MOE) was projected at: 4.3% PF resin content and 9.9% lignosulfonate addition level.

The regression equations respecting the effect of PF resin content and the lignosulfonate addition on the water absorption of the laboratory-produced MDF panels, in a coded (Equation (7)) and explicit (Equation (8)) form, are shown as follows:

$$\hat{A} = 57.66 - 0.81.X_1 - 4.11.X_2 + 0.31.X_1.X_2 + 1.51.X_1^2 + 1.61.X_2^2 \ (\%) \quad (7)$$

where $\hat{A}$ is the predicted (24 h) water absorption value (%), X_1 – PF resin content (%), X_2 – lignosulfonate addition (%)

$$\hat{A} = 102.12 - 13.49.X_1 - 2.35.X_2 + 0.06.X_1.X_2 + 1.51.X_1^2 + 0.07.X_2^2 \ (\%) \quad (8)$$

where P_f – PF resin content (%), P_l – lignosulfonate addition (%) and coefficient of determination, $R^2 = 0.98$.

Graphical representation of the above-mentioned dependences is presented in Figure 4.

Lower water absorption was clearly evidenced after increasing the PF resin content and the lignosulfonate addition. Statistically, the dependance was close to linear. As depicted in Figure 4, fibreboards meeting the European standard requirements can be produced with 3.5% PF resin and 6% lignosulfonate addition, at the lowest. At 3% PF resin level, the requirements can be met at 12.5% lignosulfonate addition, at the minimum. Within the studied variation range of the factors, it was found that the optimal 24 h water absorption value (i.e. 55.1%) can be attained at: 4.2% PF resin content and 13.1% lignosulfonate addition.

According to Dumitrescu *et al.* (2009), the addition of lignosulfonate in a PF resin results in several improvements due to the polyphenolic structure of lignin compounds and the presence of carbonyl groups, e.g. decreased adhesive viscosity, decreased reactivity, and increased pot life of the adhesive. However, up to date, the addition of unmodified lignin-based compounds into PF resins is limited to low replacement levels, because that causes reduced adhesion strength and longer pressing times, since lignin has much lower reactivity toward formaldehyde compared with phenol (Ferdosian *et al.* 2017). Hence, lignin compounds such as lignosulfonates require a chemical modification so as to increase their reactivity. For instance, sulphonation (Hemmilä *et al.* 2017) is a way for further laboratory investigation, for these types of lignosulfonate-PF resins in the future.

Conclusions

Medium-density fibreboards with acceptable physical and mechanical properties can be produced with conventional PF resin, at low addition levels, if combined with calcium lignosulfonate, specifically at 5% to 10% addition (on the dry fibres). Results from laboratory tests, with low PF resin contents (3%, 4%, 5%) and different calcium lignosulfonate additions (5% to 15%), showed that lignosulfonate-PF resin bonded MDF panels complying with the EN standard requirements can be fabricated with at least 3.5% PF resin content.

Furthermore, at the low PF resin level (3%), addition of calcium lignosulfonate should not exceed 10% to avoid unacceptable deterioration in the mechanical properties of the panels.

The use of lignin-based compounds like lignosulfonates along with PF resins is a promising approach for producing 'eco-friendly' MDF panels with acceptable properties. As a

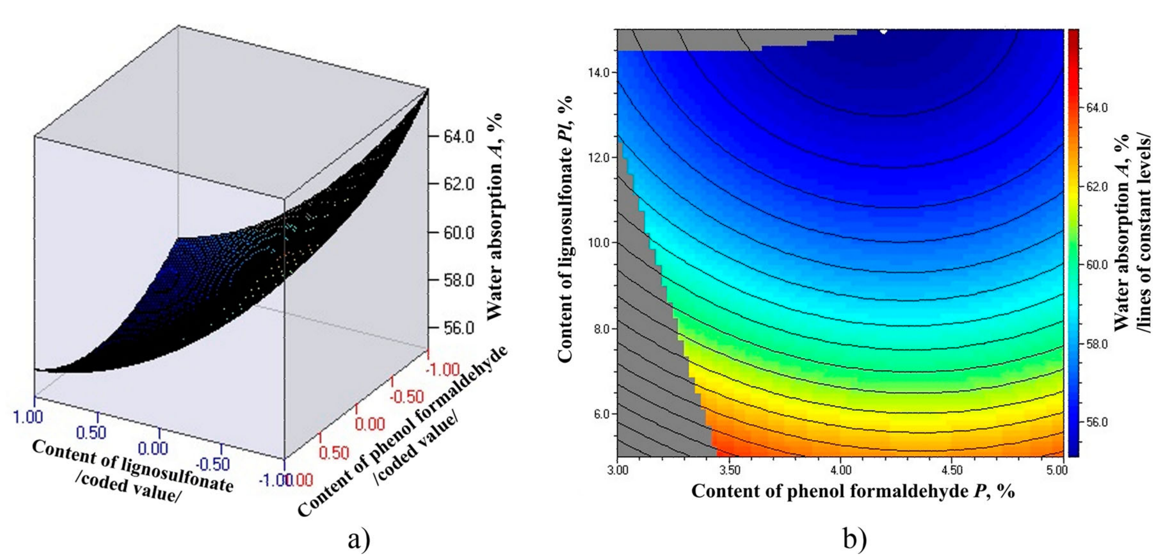


Figure 4. Variation of the water absorption of MDF panels depending upon the PF resin content and the lignosulfonate addition: (a) coded form and (b) explicit form.

result, it can be concluded that PF resin content can be reduced to 3%, with the addition of calcium lignosulfonate in the glue, even without further cross-linking.

Future studies in this topic should be aimed at lowering the PF resin content by modifying the formula of lignosulfonate additive, optimising the pressing parameters and investigating the chemical interaction between phenolic resins, lignosulfonate and wood fibres, in order to achieve an optimal performance.

Acknowledgements

This research work was supported by the project No. НИС-Б-1002/03.2019 'Exploitation Properties and Possibilities for Utilization of Eco-friendly Bio-composite Materials', which was carried out at the University of Forestry, Sofia, Bulgaria, in cooperation with the University of Thessaly (Department of Forestry, Wood Sciences & Design, Karditsa, Greece).

Disclosure statement

No potential conflict of interest was reported by the author(s).

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