Heat Transfer and Physical-Mechanical Properties Analysis of Particleboard Produced with ZnO Nanoparticles Addition

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The use of particleboards (PB) has increased quickly as an alternative engineered wood product mainly due to its having a better ratio of resistance to weight and more elimination of wood defects, such as the presence of knots. Although the panel industry has been constantly growing, innovations are still necessary to improve the final product. The use of metallic oxide nanoparticles on the wood-based panels has the potential to increase the heat transfer process and improve the physico-mechanical properties. The aim of this work was to evaluate the influence of the addition of zinc oxide (ZnO) nanoparticles in PB, correlating the physical and mechanical properties of the panel with the heat transfer process at 180 °C. The results were compared with the Brazilian standard ABNT NBR 14810-2 (2013) and the European standard EN 312 (2010), as well with works found in literature. The results showed a homogenous heat distribution during the pressing, which improved physical properties, decreasing the 24h swelling from 22.2% to 14.9% and the 24 h absorption from 30.29% to 21.0%. Besides that, MOR values was increased from 11.3 MPa to 14.5 MPa and the MOE from 1880 MPa to 2510 MPa.

Keywords: Particleboard; Physico-mechanical characteristics; Nanotechnology

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INTRODUCTION

The increasing use of wood products has prompted a more rational use of this raw material. One way that wood can be used advantageously is through the production of panels. Wood panels are produced for different applications in civil construction and furniture; one product, for example, is the particleboard (PB). Currently, PB is the most produced and most consumed wood panel in the world, representing approximately 50% of Brazil's panel production. Virtually all production is consumed by the domestic market, and PB exports are concentrated in South America, which in 2009 corresponded to 94.0% of total Brazilian PB exports (ABRAF 2013). These panel products are interesting alternatives to solid wood because in addition to their superior mechanical properties, they minimize wood defects such as knots, margins, and grain deviation, as well as the dimension limitations of wood, providing a more complete use of the raw material without compromising the quality of the final developed product.

Despite the constant improvements in wood-based panels, some features can easily be improved by nanoparticles, which have unique physical and chemical properties than the same materials at larger scales (Yu *et al.* 2008). Nanoscale materials deeply penetrate the wood, which effectively changes the surface chemistry and may result in a higher protection against moisture, better heat conduction, and an increase in the physical and mechanical properties of wood composites (Mantanis and Papadopoulos 2010; Kumar and Kim 2012).

Table 1 presents some results of the main research found on the subject with panels MDP (Medium Density Particleboard) or PB, MDF (Medium Density Fiberboard) and OSB (Oriented Strand Board), and also shows the nanoparticle used and the result obtained.

Table 1. Main Works Found on The Topic

Author	Nanoparticle	Panel	Results
Papadoulos (2010)	SurfaPoreTMW	MDP, OSB e MDF	Improved in thickness swelling.
Kumar and Kim (2012)	Aluminum oxide (Al_2O_3)	MDF	Tendency of improvement of physical and mechanical properties and greater workability of the panel.
Salari *et al.* (2013)	Nanoparticles of Sílica	OSB	Improved of Internal bonding and heat conduction.
Taghiyari and Nouri (2015)	Nano-wollastonite (NW)	MDF	Improved water absorption properties and thickness swelling.
Rangavar and Hoseiny (2015)	Nanoparticles of copper	MDP	Increased MOR value and improved internal bonding.
Cardoso *et al.* (2016)	Nanocellulose	MDP	Improvement of physical properties.

Regarding heat transfer in particulate panels, new probabilities have arisen with the use of nanoparticles. The addition of metallic nanoparticles in the resin increases the thermal conductivity of the wood panel, which can shorten the pressing time (Kumar *et al.* 2013).

Heat transfer plays an important role for the resin curing temperature in wood-based panels, acting from the surface of the panel to its center through the migration of the vapor. Thus, studying this phenomenon may contribute to the improvement of certain properties of the panel.

Some studies show that nanoparticles intensify the heat transference. Anoop *et al.* (2009) investigated the effect of different nanoparticle sizes on the heat transfer, observing that the nanofluids have high heat transfer capacity. Mohammed *et al.* (2018) used ZnO nanoparticles on wood composites to improve weather resistance and better physical and mechanical properties. In addition, Bak and Németh (2018) verified that zinc oxide provides effective protection against fungi, showing good protection for both investigated fungi, *Coniophora puteana* and *Coriolus versicolor*. These studies indicate that the use of nanoparticles in wood-based panels can be an interesting alternative to improve the heat transfer as well as the physic-mechanical properties of the panels.

The good resin fixation is responsible for appropriate properties of panels, so if a good heat transfer was achieved, then, the adhesive fixation is will be better. So, the ZnO can improves the heat transfer and the interaction between wood particles and resin whether the good adhesive fixation was achieved.

Thus, aiming at a sustainable production, with lower temperature and pressing time, in this paper, the addition of zinc oxide nanoparticles, produced with a different method, a sol-gel process was examined to improve the performance of PB panels produced with urea-formaldehyde, especially in contact with water. This process is a promising method to get nanostructured powders in several ways, being a technological advance for panels and reveal an interesting alternative for the industry.

EXPERIMENTAL

Particleboard Production

The particles used to produce the panels were wood of *Eucalyptus Urophila x Grandis* hybrid with dimensions of the particles sieved 9, 16, 35 and 60 mesh, donated by a timber company (Botucatu, Brazil). These particles were dried in an oven at 103 °C ± 2 °C, until 3% moisture content was obtained.

The adhesive used was urea-formaldehyde (UF) with a solids content of 66%; its dosage was fixed based on the particles weight using 8% of adhesive for the internal layer and 10% of adhesive for the external layer. The catalyst contained ammonium sulfate and an emulsion of paraffin, with a solids content of 13.10% and 57.2%, respectively. The panels were produced in two samples, without the addition of nanoparticles (T1) and with the addition of nanoparticles (T2). For the sample T2, 1% of ZnO nanoparticles were added. The nanoparticles were synthesized by a sol-gel method (Favarim and Leite 2018).

For the particle mattress formation, 2000 g of wood particles were used in the proportion of 20% to 60% to 20%, where 1200 g was for internal layer (9 and 16 mesh), 400 g for top layer and 400 g for bottom layer (35 and 60 mesh). The nanoparticles were added to the adhesive, and it was sprayed by an air spray gun onto the wood particles while they were mixed, ensuring good homogeneity. In all 12 panels of 45 x 45 cm were produced.

Heat Transfer

For the heat transfer study, a thermocouple type K was used, attached with an automated data acquisition system. The thermocouple was inserted into the internal layer of the mattress to measure the temperatures during the pressing.

After the particle mattress formation, it was pre-pressed at room temperature for 300 s, and pressure of 0.3 MPa. A hot pressing with temperature of 180 °C and pressure of 4 MPa, resulting from the 235 bar hydraulic pressure applied to the pressure gauge, was performed with a total cycle of 600 s, where was divided into three cycles of 3 minutes with 30 s break.

Physical and Mechanical Tests

The physical tests were performed to determine the density, moisture content, swelling in thickness, and water absorption after 2 h and 24 h immersion. To determine the mechanical properties, a bending test was performed to determine the modulus of elasticity (MOE) and the modulus of rupture (MOR). For the tests, 10 specimens were used for each one, as indicated by Brazilian standard ABNT NBR 14810-2 (2013).

Determination of apparent density

The density for each sample was determined using ten square specimens with 50 mm sides and was calculated using Eq. 1. The width and length were measured with a

caliper, the thickness was measured in five different points (four corners and at the center) with a micrometer within 0.001 mm of precision, and the mass was determined using a precision scale,

$$D = \frac{\mathrm{m}}{\mathrm{w} \times \mathrm{l} \times \mathrm{t}} \times 1{,}000{,}000 \quad (1)$$

where D is the apparent density (kg × m^{-3}), m is the mass of the specimen (g), w is the width (mm), l is the length (mm), and t is the thickness (mm).

Determination of moisture content

Each sample moisture content was determined using ten square specimens with 50 mm sides and their initial mass was determined with a precision scale. Samples were dried in a laboratory oven at 103 °C ± 2 °C until they reached a constant mass (*i.e.*, the fluctuation between mass measurements was less than 0.1 %). The sample moisture content was calculated by Eq. 2,

$$U = \frac{m_i \times m_d}{m_i} \times 100 \quad (2)$$

where U is the moisture content (%), m_i is the initial mass of the specimen (g), and m_d is the dried mass of the specimen (g).

Determination of thickness swelling and absorption

Ten square specimens with dimensions of 50 mm × 50 mm were completely submerged in water to determine the thickness swelling and absorption. The thickness swelling was determined by observing any increase in the thickness dimensions after the specimens were soaked in water for 24 h, measured by a micrometer with a precision of 0.001 mm. The water had a constant temperature of 25 °C and a pH of 7 during the test. The same procedure was carried out to determine the absorption. The thickness swelling and the absorption were determined using Eqs. 3 and 4, respectively,

$$TS = \frac{t_s \times t_i}{t_i} \times 100 \quad (3)$$

$$A = \frac{a_s \times a_i}{a_i} \times 100 \quad (4)$$

where TS is the thickness swelling (%), t_s is the specimen thickness after water soaking (mm), and t_i is the initial thickness (mm); A is the thickness absorption (%), a_s is the absorption after the water soaking (mm), and a_i is the initial absorption (mm).

Determination of elasticity modulus and rupture modulus

The bending test was used to obtain the modulus of elasticity (MOE) and modulus of rupture (MOR). The test was performed on a universal testing machine (EMIC Hidralmac, Araraquara, Brazil), with a sample placed over two supports and with the distance between supports being 20 times the thickness of the specimen, as shown in Fig. 1.

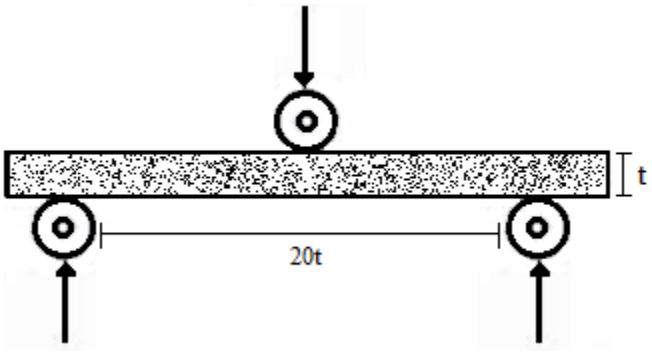

Fig. 1. Distance between supports adapted from ABNT NBR 14810-2 (2013)

The MOR values were obtained using Eq. 5, and the MOE values were obtained with Eq. 6. These equations were added to the software that accompanies the universal testing machine. Equation 5 is shown below,

$$MOR = \frac{1.5 \times L \times D}{w \times t^2} \quad (5)$$

where *MOR* is the modulus of rupture (MPa), *L* is the read rupture load (N), *D* is the distance between supports (mm), *W* is the specimen width (mm), and *t* is the average thickness taken at three points on the specimen (mm). Equation 6 is shown below,

$$MOE = \frac{L_1 \times D_3}{4 \times d \times W \times E_3} \quad (6)$$

where *MOE* is the modulus of elasticity (MPa), L_1 is the load on the proportional limit (N), *D* is the distance between the supports of the appliance (mm), *d* is the deflection corresponding to the load P_1 (mm), *W* is the specimen width (mm), and *E* is the average thickness taken at three points on the specimen (mm).

Internal bonding - Perpendicular traction

The internal bonding test was performed with a perpendicular traction of the panel, where the specimen was assembled with a 50 mm × 50 mm controlled velocity extension. The calculation of the resistance was done perpendicularly through Eq. 7, and that was added to the software used with the universal testing machine. Equation 7 is shown below,

$$TP = \frac{L}{S} \quad (7)$$

where *TP* is the perpendicular tensile strength (MPa), *L* is the burst load (N), and *S* is the surface area of the specimen (mm²).

Statistical Analysis

Statistical analyzes were performed with R version 3.2.0 software (www.r-project.org), by Test T with significance levels of 5%. It was tested in two samples, the sample 1 (T1) had no added nanoparticles, and sample 2 (T2) had 1% of nanoparticles added. After the tests, the results were compared with the test standards ABNT NBR 14810 (2013) and EN 312 (2010).

RESULTS AND DISCUSSION

Physical Properties

Tables 2 to 7 presents the average results obtained for the physical tests of density, moisture content, thickness swelling, and absorption for all specimens. The results were within the normative specifications proposed by the Brazilian standard - ABNT NBR 14810 (2013) and the European standard (EN) - EN 312 (2010).

Table 2. Average Density (Kg/m³)

Condition	T1	T2	ABNT	EN
	0%	1%	0%	0%
Mean[1]	599.96	676.57	551 - 750	500 - 800
SD[2]	67.41	50.34	-	-
(%CV) [3]	11.23	7.44	-	-
P-value	0.000675457			

* [1] Means (Test t, α = 0.05); [2] Standard deviation of the data; [3] Coefficient of Variance (%CV)

The average density increased after the addition, and with P-value 0.0067 was obtained statistical difference for Test T, with 5% significance. This occurred because of a decreased average thickness. Therefore, the higher density of the panels, especially the external layers, in T2 was due to the acceleration of the heat transfer provided by the nanomaterials, which provided a smaller thickness and increased the density.

Table 3. Moisture Content (%)

Condition	T1	T2	ABNT	EN
	0%	1%	0%	0%
Mean[1]	6.45	6.61	5 - 11%	-
SD[2]	0.51	0.58	-	-
(%CV) [3]	7.91	8.77	-	-
P-value	0.4507043			

* [1] Means (Test t, α = 0.05); [2] Standard deviation of the data; [3] Coefficient of Variance (%CV)

For moisture content, with P-value 0.45, there was no statistical difference between the samples for Test T with 5% significance. Therefore, it was concluded that the moisture content did not interfere with the other properties. When compared with other studies, the panels examined in this study remained in the same mean values. Valle (2015) studied PB with the addition of silica (SiO_2) nanoparticles and found an average density value of 866 kg/m³ and moisture content values of 6.95% and 6.46% for PB panels without nanoparticles and 4% of SiO_2, respectively, similar values for this work. Cardoso *et al.* (2016) obtained moisture content values of 9.7% and 10.8% with the addition of 2% of nanocellulose and 5% of nanocellulose, respectively, in agglomerated panels made with urea-formaldehyde resin. The use of ZnO nanoparticles allowed for values that were in accordance with the Brazilian standard during the 2 h and 24 h swelling tests. However, the T1 panels presented higher swelling for both tests with statistical difference in Test T and 5% significance. Roumeli *et al.* (2012) and Valle (2015) both treated the PB panels with silica nanoparticles. Both studies show an improvement for the swelling during the 24 h test, obtaining values of 29% and 27% and 62% and 36%, respectively, for panels without and with the addition of the nanomaterial.

Table 4. 2h Thickness Swelling (%)

Condition	T1	T2	ABNT	EN
	0%	1%	0%	0%
Mean[1]	10.60	4.30	< 8%	<12%
SD[2]	1.60	1.03	-	-
(%CV) [3]	15.09	23.95	-	-
P-value	0.04202537			
* [1] Means (Test t, α = 0.05); [2] Standard deviation of the data; [3] Coefficient of Variance (%CV)				

Table 5. 24h Thickness Swelling (%)

Condition	T1	T2	ABNT	EN
	0%	1%	0%	0%
Mean[1]	22.20	14.90	<18%	<16%
SD[2]	1.70	1.50	-	-
(%CV) [3]	7.66	10.07	-	-
P-value	0.01822358			
* [1] Means (Test t, α = 0.05); [2] Standard deviation of the data; [3] Coefficient of Variance (%CV)				

Table 6. 2h Absorption (%)

Condition	T1	T2	ABNT	EN
	0%	1%	0%	0%
Mean[1]	9.24	5.78	-	-
SD[2]	1.60	1.25	-	-
(%CV) [3]	17.31	21.62	-	-
P-value	0.02190089			
* [1] Means (Test t, α = 0.05); [2] Standard deviation of the data; [3] Coefficient of Variance (%CV)				

Table 7. 24h Absorption (%)

Condition	T1	T2	ABNT	EN
	0%	1%	0%	0%
Mean[1]	30.29	21.01	-	-
SD[2]	3.35	1.17	-	-
(%CV) [3]	11.06	5.57	-	-
P-value	0.00654103			
* [1] Means (Test t, α = 0.05); [2] Standard deviation of the data; [3] Coefficient of Variance (%CV)				

Similarly as thickness swelling, the absorption test presented good values for the panels treated with ZnO nanoparticles. When comparing the T1 and T2 for the 2 h water absorption test, there was an approximate decrease of 50%. Statistical difference was also obtained in the Test t, with 5% of significance.

Despite the absence of normative data for the absorption test, recent studies show verified higher values for both the 2 h and 24 h absorption tests. Surdi (2015) has data that reaches approximately 66% and 84%, making use of eucalyptus residues for 2 h and 24 h tests, respectively. Cardoso *et al.* (2016) has data that reaches 180% and 220%, making use of 2% of nanocellulose and 5% of nanocellulose for the 24 h absorption tests.

These results reinforce the quality of the produced panel in this study as well as demonstrate that the use of ZnO nanoparticles may better cure the resin, which increased the internal adhesion among the particles of the panel, preventing higher water absorption and swelling in PB panels.

Mechanical Properties

Table 7 to 9 presents the average results obtained for the mechanical tests of Modulus of Rupture, Modulus of Elasticity and Internal Bond, for all specimens. The results were within the normative specifications proposed by the Brazilian standard - ABNT NBR 14810 (2013) and the European standard (EN) - EN 312 (2010).

Table 8. Modulus of Elasticity – MOE (MPa)

Condition	T1	T2	ABNT	EN
	0%	1%	0%	0%
Mean[1]	1878.25	2511.28	> 1800	> 1800
SD[2]	292.36	223.52	-	-
(%CV) [3]	15.56	8.90	-	-
P-value	0,04943976			
* [1] Means (Test t, α = 0.05); [2] Standard deviation of the data; [3] Coefficient of Variance (%CV)				

The Brazilian standard - ABNT NBR 14810 (2013) and the European standard (EN) - EN 312 (2010) specify a minimum value of 1800 MPa for the modulus of elasticity (MOE), considering non-structural panels with a thickness of 6 mm to 13 mm. All samples were in accordance with the standards; however, for T2, the MOE values increased more than 20% and obtained statistical difference for Test T with 5% of significance.

Table 9. Modulus of Rupture – MOR (MPa)

Condition	T1	T2	ABNT	EN
	0%	1%	0%	0%
Mean[1]	11.32	14.52	> 11	> 11
SD[2]	2.08	1.56	-	-
(%CV) [3]	18.37	10.74	-	-
P-value	0,01215903			
* [1] Means (Test t, α = 0.05); [2] Standard deviation of the data; [3] Coefficient of Variance (%CV)				

All samples tested for rupture stress satisfied both ABNT NBR 14810 (2013) and EN 312 (2010). The modulus of rupture (MOR) for the ABNT NBR 14810 (2013) indicates a minimum value of 11 MPa, while the MOR for the European standard is 11.5 MPa. The addition of ZnO nanoparticles increased the MOR values, which increased the performance of the PB panels.

The mean values for the MOR of the different samples presented a statistical significant difference with 95% confidence, where T2 presented better results. Valle (2015) found MOR values close to 12.2 MPa. Taghiyari (2011) studied PB panels with the addition of copper nanoparticles and found MOR values between 11.6 MPa and 12.2 MPa, which agrees with the values found in this research.

Table 10. Internal Bond (MPa)

Condition	T1	T2	ABNT	EN
	0%	1%	0%	0%
Mean[1]	0.44	0.46	> 0.4	> 0.4
SD[2]	0.06	0.05	-	-
(%CV)[3]	13.64	10.87	-	-
P-value	0,8985573			

* [1] Means (Test t, α = 0.05); [2] Standard deviation of the data; [3] Coefficient of Variance (%CV)

The analysis of the internal bond obtained by the perpendicular traction test for the produced panels presented similar values and had no statistical difference between them with Test t and 5% of significance. All the obtained results were in accordance with the values established by the Brazilian standard and the European standard, as well as other published papers. Taghiyari (2011) added copper nanoparticles to PB panels and obtained approximate values of 0.48 MPa.

The performance increased with the addition of 1% of ZnO nanoparticles to the production of PB panels when analyzing the MOR, MOE, and internal bonding.

Heat Transfer

The pressing temperature of the panels was analyzed during a hot pressing by a type k thermocouple because curing the resin could have been affected by the addition of the ZnO nanoparticles. As shown in Fig. 2, there was an evolution of the temperature during the pressing process for both T1 (0%) and T2 (1%).

The addition of ZnO nanoparticles on the PB panel induced less variation on the temperature over time during the hot pressing, as shown by the T2 line. T1 slowly increased the temperature and presented some oscillation. The final temperature for T2 was obtained at approximately 220 s, while the T1 needed approximately 380 s to reach its final temperature.

As shown in Fig. 1, the addition of nanoparticles accelerated the heating and curing process of the panel in the innermost region. In the first cycle (up to 180 s), T2 had already reached 100 °C while the T1 during this period was still approximately 85 °C. The curing urea-formaldehyde (UF) temperature should be between 95 °C and 130 °C (Iwakiri 2005).

Achieving the needed cure temperature while in the first cycle provided better densification and material compacting, which resulted in a higher density and smaller voids. The higher density and smaller voids improved the panels performance when in contact with water, reducing the swelling in thickness and the water absorption. For the mechanical properties, the higher densification provided better mechanical properties, especially in the strength results and the rigidity of the panels, obtained from the bending test. For the internal bond test, there was no significant statistical difference because the final cure temperature was approximately the same for both samples.

The graph presented on Fig. 2 shows that the internal temperature for T2 was reached at 180 s and remained nearly constant up to 600 s. These results suggest that future studies use reduced press cycles, aiming for less pressing time, generating increased production, and lower energy consumption. Lima *et al.* (2018) indicate that pressing times between 400 s and 600 s do not present differences in the physical and mechanical properties of the panels. Additionally, the nanoparticles' association can further reduce this pressing time without compromising the final technical performance of the panel.

Although the highest temperature on the plates of the press was 180 °C, the

maximum temperature reached within the panels was approximately 120 °C for both panels. However, T2 constantly increased the temperature, prevented major oscillations, reached its final temperature way before T1, and accelerated the resin cure.

Simple changes and with low-cost might allow for a shorter pressing time, a decreased pressing temperature, or both, keeping the same quality of the PB panels. Similar results were achieved by Kumar *et al.* (2013), with the addition of Al_2O_3 nanoparticles in wood panels that verify an increase in the panel properties.

By improving the temperature distribution in the PB panels during the pressing, with the addition of the ZnO nanoparticles, it was possible to study different ways of optimizing the pressing cycle variables, allowing for both a shorter pressing time and a decrease in the pressing temperature. In addition, the wood particles and the resin, which define the physical and mechanical properties, had better interactions with each other.

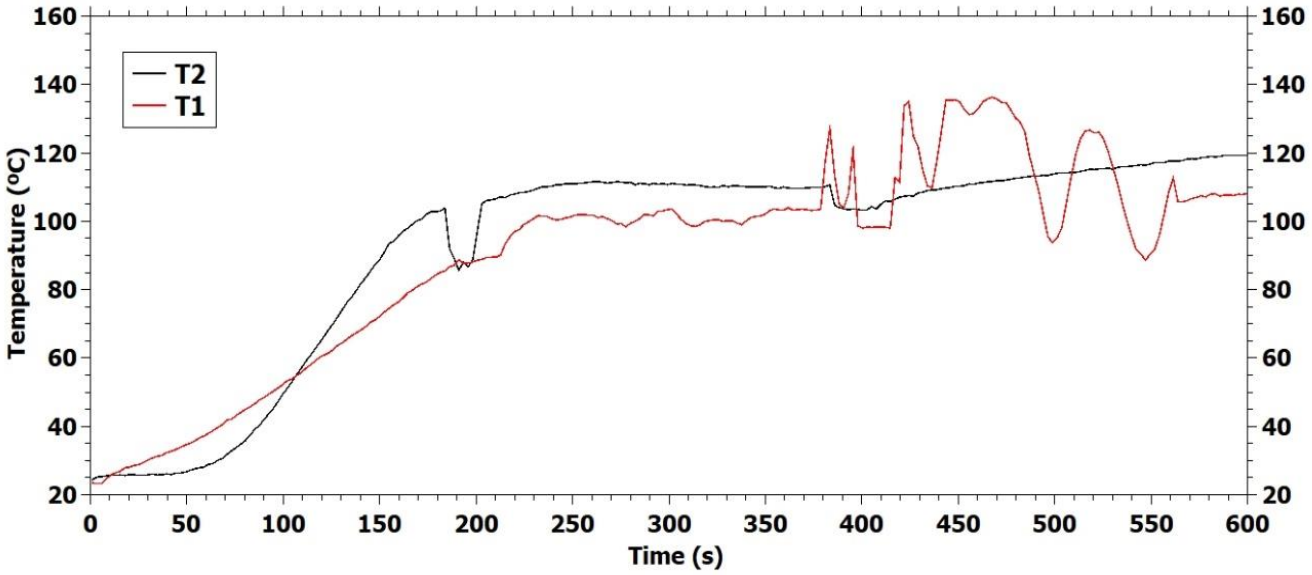

Fig. 2. Curves of temperature with addition (T2) and no addition (T1) of nanoparticles in core layer during pressing

CONCLUSIONS

1. The addition of 1% of ZnO nanoparticles to particleboard (PB) showed an increase in the mechanical and physical properties, especially for swelling and water absorption. These are considered critical properties for PB panels.
2. Despite the T1 and T2 reaching the same final temperatures, the treated panels (T2) gradually increased in temperature, reaching its final temperature before the non-treated panels (T1). This phenomenon accelerated the cure of the resin, allowing for a better interaction between the wood particles and improving the physic-mechanical properties of the panels.
3. Since the heat transfer was improved and the good adhesive fixation was achieved, a better interaction between wood particles and the resin was achieved. This characteristic also improved the water absorption and swelling because empty spaces between wood particles were smaller.

4. Using a small amount of ZnO nanoparticles improved the quality of the panel. This was a simple and low-cost implementation and is promising when considering the thermal energy efficiency and lower production costs.

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