

THE EFFECT OF ADDING GRAPHENE OXIDE TO UREA FORMALDEHYDE RESIN AND ITS EFFICACY ON THREE-LAYERED PARTICLEBOARD

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ABSTRACT

The research for improving adhesives and the properties of wood composites is always important for industry. The aim of this study is investigating the effect of adding graphene oxide in the UF resin on the functional properties of particleboard panels with an average thickness of 16 mm. The influence of graphene oxide content (0; 0,25 %; 0,5 % and 0,75 % based on the dry weight of the UF resin) and pressing time (4 and 5 minutes) on the internal bonding, bending strength, modulus of elasticity and dimensional stability were studied. The results showed that the use of graphene oxide in the UF resin caused an improvement of the mechanical and physical properties of the particleboards. Also, comparing two different pressing times, the boards made by 4 minutes with graphene oxide is preferred without negative effect on the functional properties of particleboards.

Keywords: Particleboard, dimensional stability, graphene oxide, internal bonding, Wood composites

INTRODUCTION

Graphene is a carbon monolayer material arranged into a honeycomb lattice (Balasubramanian and Chowdhury 2015) that has proved itself as an exciting material with opportunities for new scientific knowledge because of its especial electrical, thermal and mechanical properties related to its two dimensional (2D) structure that offers great potential for several applications (Tadyszak *et al.* 2018). Graphene has also emerged as good non-organic filler for composites, improving their properties (Dreyer *et al.* 2010, Allen *et al.* 2010, Kuilla *et al.* 2010, Bose *et al.* 2010).

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Graphene oxide (GO) is one of the promising derivatives of graphene. It is a chemically modified graphene that has a similar hexagonal carbon structure to graphene and contains oxygen functional groups, such as carboxyl (-OOH), hydroxyl (-OH), or epoxy (-O) (Figure 2) (Tadyszak *et al.* 2018). Microscopic and spectroscopic measurements demonstrate that GO preserves the honeycomb lattice of graphene, whereas the carbon sheet is distorted due to the chemisorbed oxygenated groups. Graphene oxide inherits the better mechanical strength of graphene and in order to its unique interlayer structure, shows high stiffness and failure resistance (Huang *et al.* 2020). But these oxygenated groups on its structure are responsible for many advantages over graphene, including the possibility for surface functionalization which have made it suitable for use in nanocomposite materials (Smith *et al.* 2019). The mechanical properties of graphene oxide can be effectively adjusted by controlling the concentration and species of oxygenated groups (Huang *et al.* 2020). Furthermore, due to the presence of oxygen functionalities, graphene oxide can easily disperse in organic solvents, water, and different matrixes (Tadyszak and *et al.* 2018, Stankovich *et al.* 2007). Therefore it has higher solubility than graphene (Kuilla *et al.* 2010). This is a main benefit in order to mixing the material with polymers to increase their mechanical and electrical properties (Perrozzi *et al.* 2015). Graphene oxide flakes can be used in different forms, such as layers, fibers, or three-dimensional foams (Tadyszak and *et al.* 2018).

So, because of adjustable nature of graphene oxide, which presents controllable electronic, mechanical and thermal properties, it can be used as a material for nanomechanical systems and nanocomposites to improve new products (Smith *et al.* 2019). Sarkar and Kim (2016) investigated the synthesis of graphene oxide-epoxy resin encapsulated urea-formaldehyde microcapsule. They stated that the microcapsules showed greater thermal stability with increasing the GO content. Increasing thermal stability may be useful for materials used in high temperature. So, various researches demonstrate that polymer nanocomposites benefit greatly from the use of graphene oxide and its derivatives as fillers (Kim *et al.* 2010). In this regard Yadav *et al.* (2013) introduced graphene oxide as nano filler for the production of biodegradable nanocomposites made of carboxyl methyl cellulose (CMC). They showed that a strong adhesion is formed between graphene oxide and CMC through the formation of hydrogen bonds which led to improved mechanical properties. Also, Cheng-An *et al.* (2017) showed the effects of graphene oxide in polyvinyl alcohol (PVA) films. They found increasing the tensile strength of the nanocomposite up to 59,6 MPa more than the pure PVA film, by using 20 % graphene oxide. They reported that the improvement in mechanical properties is related to the strength of the graphene oxide filler and the matrix/filler interface and a high degree of hydrogen bonding is formed between the OH groups of PVA and oxygen functionalities of the graphene oxide.

Urea formaldehyde (UF) resins are important in wood composite industry especially particleboards for indoor uses and are used in a broad range because of their low cost, high reactivity and simple processing. But one of the important disadvantages of this adhesive in particleboard production is its low resistance to heat and humidity and the release of formaldehyde. In this regard the addition of particulate fillers like soy protein (Qu *et al.* 2015), corn flour (Moubarik *et al.* 2013), furfural (Esmaeili *et al.* 2017), starch (Warui Kariuki *et al.* 2019) and liquefied wood waste (Janiszewska *et al.* 2016) and etc., to UF resin is of interest in order to improve the functionality of this resin and reduce its consumption (Pinkl *et al.* 2018) due to the reduction of formaldehyde emissions and consequently the reduction of environmental problems. Furthermore, using different additives may limit the physical and mechanical properties of the boards. Therefore, it seems necessary to find a suitable material that can improve the properties of urea-formaldehyde resin and functional properties of particleboards. Thus, considering the GO especial properties, the incorporation of it within UF resin can be a possible method to improve the functional properties of particleboard, which would be beneficial to extend its applicability.

MATERIALS AND METHODS

Raw materials

For the production of laboratory particleboards, the coarse industrial wood particles (for the middle layer of composite) and fine industrial wood particles (for the surface layer of composite) were provided by commercial particleboard plant in Iran. Urea formaldehyde resin was supplied by Samed Mashhad Glue Co. in Iran. Also, Ammonium Chloride powder was used 2 % based on the dry weight of the UF resin as a hardener for UF resin. Specified graphene oxide powder was obtained from the API Co. With the black color, the number of impurities <70 ppm, the ratio of carbon to oxygen: 8, dimensions of graphene sheets <44 μm and the thickness of graphene sheets are less than 3 nm.

Particleboards manufacturing

In order to make three layered particleboards, the wood particles were dried to a moisture content of 5 % (for the core layer) and 8 % (for the surface layers) inside in a laboratory dryer with a gentle temperature of 80 °C and instant control. Wood particles were uniformly sprayed with a commercial liquid UF resin (50 % resin content, pH 8, specific gravity 1,28 viscosity 450) that was mixed with graphene oxide powder by a cylindrical resin blender. The resin content was 8 % for the core layer and 10 % for the surface layer based on the oven dry weight of wood particles and mixed with graphene oxide powder as a variable with 0 % (control) 0,25 %; 0,5 % and 0,75 % based on the dry weight of the UF resin. Composite mats were manufactured manually using a 400 mm × 400 mm × 16 mm forming box (Figure 6) with a target density of 700 kg/m³. In this study no wax was added to the mixtures. Board consolidation was performed by pressing the particle mat between two aluminum plates using a hydraulic press under 30 kg/cm² pressure at temperature of 160 °C for 4 and 5 minutes (pressing time was also as variable). The thickness of the boards was controlled by a spacer with a thickness of 16 mm (Figure 1). Three replicate particleboards were made for each treatment combination. After pressing, the boards were conditioned for 10 days at 21 °C and 65 % relative humidity prior to physical and mechanical testing. After the desired time, the boards were trimmed and cut for physical and mechanical tests. Internal bonding (IB) according to EN 319 (1993), bending strength (MOR) and modulus of elasticity (MOE) according to EN 310 (1993) and thickness swelling (TS) of the boards within 2 and 24 hours immersion in water at 20 °C according to EN 317 (1993) were measured. Table 1 presents the dimensions of test samples.



Figure 1: Preparation of particle mat (a) and board consolidation by pressing the mat using hydraulic press (b).

Table 1: The dimensions of the test samples

Experimental sample	Length (mm)	Width (mm)	Thickness (mm)
IB	50	50	16
MOR & MOE	370	50	16
TS	50	50	16

RESULTS AND DISCUSSION

Mechanical properties

The mechanical properties especially internal bonding of the particleboards is the main factor in the assessment of the feasibility of the manufacturing technique. Figure 2 shows the internal bonding values obtained from particleboards made in this study. Differences were found in the values of internal bonding under effect of various graphene oxides content and pressing time. Figure 2 show that the addition of graphene oxide improved the internal bonding of the boards. It presents an increasing trend by increasing the GO content and achieves a high value at the GO content of 0,75 %. It demonstrates that the control samples without graphene oxide in its structure showed lower internal bonding values compared to other boards that have graphene oxide in their structures. Due to the presences of hydroxyl groups (-OH) in the graphene oxide structure, it is

possible that an adhesion is formed through the formation of hydrogen bonds between graphene oxide and target substrate and this connection can be led to improved mechanical properties (Yadav *et al.* 2013) which might be occurred between graphene oxide and UF resin functional groups too. Furthermore, pressing time has a considerable effect on bonding performance of the boards. As can be seen from figure 2 the particleboards made by pressing time of 4 minutes showed lower internal bonding. Whereas the boards made by higher pressing time of 5 minutes showed a remarkable increase in the IB of the boards. Particleboard composites containing 0,75 % graphene oxide and pressed for 5 minutes showed the highest internal bonding. However, as shown in Figure 2, the internal bonding value of all treatments increased compared to the control. This is true even for treatments that made with a pressing time of 4 minutes. It means that composites including different amounts of graphene oxide and pressed for 4 minutes also have more internal bonding than the control. Balandin *et al.* (2008) states that the graphene oxide has a high thermal conductivity and the latter result may refer to increasing the speed of heat transfer to the middle layer of the particleboards by using graphene oxide which resulted in improved adhesion between wood particles. Also, looking at Figure 2, it can be seen that the internal bonding of all particleboards, even boards pressed under 4 minutes, is more than the EN standard. According to the European standard, the amount of internal bonding of particleboard as construction boards for interior use (P1), as furniture boards for interior use (P2) and as non-loaded-bearing use, a board that withstands moisture better than standard particleboard (P3) is 0,24; 0,35 and 0,45 MPa respectively. But as the Figure 2 shows, the internal bonding of all the boards in this research is more than 0,5 MPa. It is to be noted that even the internal bonding of the boards pressed for 4 minutes with the use of graphene oxide has increased compared to the control sample. This means that the lowest amount of graphene oxide can be used to make particleboards with a shorter pressing time, while their internal bonding is nearly doubled. That means, with the minimum amount of graphene oxide and the minimum pressing time used in this research, can be made the boards with much more internal bonding than the control, P1, P2 and P3. The results of this study show that by consuming a low amount of graphene oxide, it is possible to reduce the pressing time which is one of the important factors in the production line and also to make boards with high strengths and suitable for various uses (P1, P2 and P3).

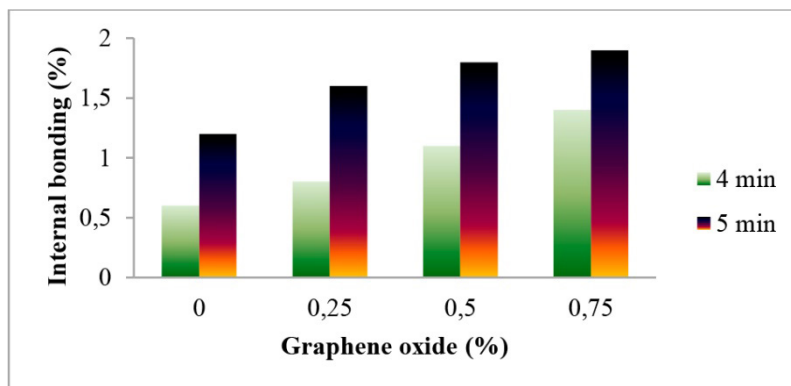


Figure 2: Internal bonding as a function of graphene oxide content and pressing time.

Figure 3 and Figure 4 present the results of bending strength (MOR) and modulus of elasticity (MOE) of the particleboards. Both MOR and MOE showed positive changes by addition of graphene oxide and reducing pressing time. But as is clear the MOE is increased by adding the graphene oxide to the boards because of its high young's modulus (Suk *et al.* 2010, Liu *et al.* 2012, Gao *et al.* 2011, Lee *et al.* 2008) and also the addition of different levels of graphene oxide had improvement the flexibility of particleboard and increased MOR. A high value of MOR was obtained at the GO content of 0,75 % and the pressing time of 4 and 5 minutes. Some research recorded that graphene oxide can be added into other materials to improve their bending mechanics. According to Poulin *et al.* (2016) graphene oxide is very flexible under bending and the flexibility of GO can be 100 times higher than the graphene. Also, Cheng *et al.* (2016) added GO to polymer composites consisting of surfactant-wrapped/doped polyaniline (PANI) and divinylbenzene (DVB). They recorded that the bending strength represents an increasing trend by increasing the GO content and achieves a high value of 46,6 MPa at the GO content of 0,6 %. Also they showed that the bending modulus increases by adding the graphene oxide even with only 0,45 % and bending modulus of the composites exhibits at least 29,8 % increment.

Figure 3 and Figure 4 demonstrate MOR and MOE of the particleboards with a reverse trend with reducing pressing time. But as can be seen from Figure 3 and Figure 4, with decreasing pressing time, MOR and MOE were decreased but they were still above the standard limit. As a result, due to the importance of pressing time

in the production cost of particleboards, less pressing time can be used to make them without reducing the MOR and MOE below the standard limit. The European standard reports that the amount of modulus of rupture of particleboard as construction boards for interior use (P1), as furniture boards for interior use (P2) and as non-loaded-bearing use, a board that withstands moisture better than standard particleboard (P3) is 11,5; 13 and 14 MPa respectively and the amount of modulus of elasticity of particleboard as furniture boards for interior use (P2) and as non-loaded-bearing use, a board that withstands moisture better than standard particleboard (P3) is 1600 and 1950 MPa respectively. According to the results, the MOR and MOE resistance of all treatments are higher than the values mentioned in this standard. Depending on the Figure 3 and Figure 4, it can be inferred that by consuming a low amount of graphene oxide, the pressing time can be reduced to 4 minutes, while the MOR and MOE strength of the boards are much higher than the EN standard. As a result, by consuming the minimum amount of graphene oxide studied in this research, it is possible to make standard boards without need of increasing the pressing time up to 5 minutes.

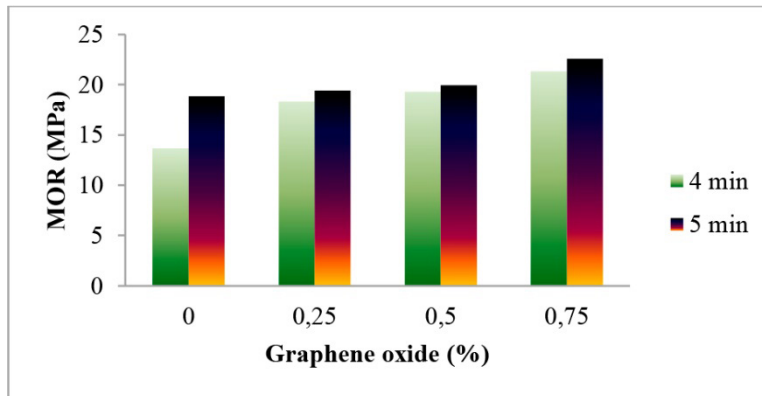


Figure 3: Bending strength as a function of graphene oxide content and pressing time.

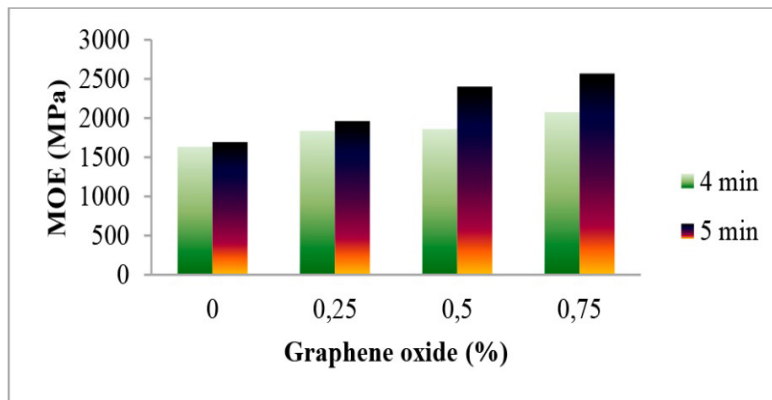


Figure 4: Modulus of elasticity as a function of graphene oxide content and pressing time.

Physical properties

Figure 5 and Figure 6 show the decreasing trend of thickness swelling (TS) of particleboards by arising graphene oxide content and pressing time. The graphene oxide amount and pressing time would affect the thickness swelling of particleboards within 2 and 24 hours immersion in water. It is obvious from Figure 5 and Figure 6 that adding graphene oxide content and increasing pressing time led to considerable decrease of TS in particleboards. The minimum TS occurred in particleboards containing 0,75% graphene oxide and pressed for 5 minutes. Also the control sample without any graphene oxide which has been pressed for 4 minutes showed the maximum TS. According to the European standard, the amount of thickness swelling of particleboard after 24 hours immersion in water as non-loaded-bearing use, a board that withstands moisture better than standard particleboard (P3) is 14 percent. But the important thing is that, as can be seen in Figure 6, all particleboards

containing graphene oxide and pressed under 4 and 5 minutes have the EN standard of TS. Figure 6 illustrates that adding graphene oxide led to decrease in TS in particleboards even by reducing the pressing time to 4 minutes. Kuila *et al.* (2012) reported that the addition of functionalized graphene in the nanocomposites increased its dimensional stability due to increased material hardness. This can be considered as a good achievement to reduce the pressing time.

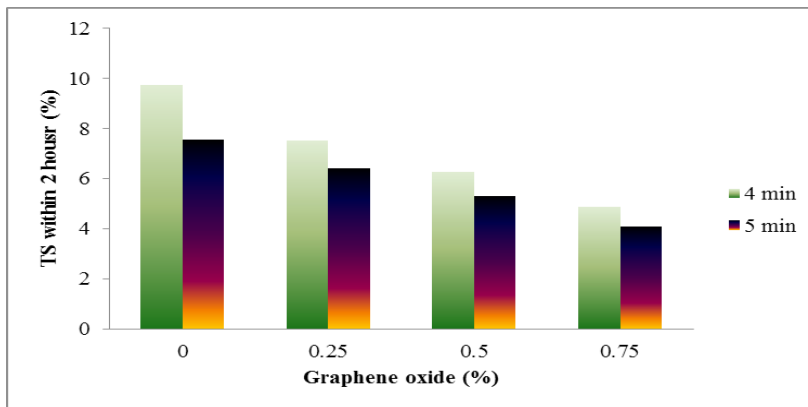


Figure 5: Thickness swelling within 2 hours immersion in water as a function of graphene oxide content and pressing time

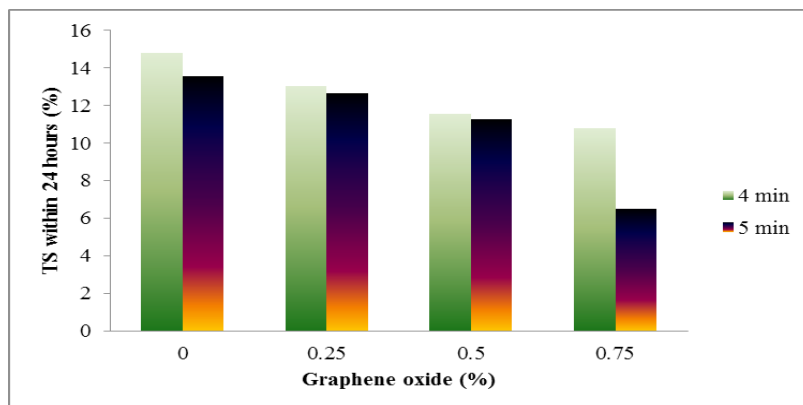


Figure 6: Thickness swelling within 24 hours immersion in water as a function of graphene oxide content and pressing time.

CONCLUSIONS

High performance particleboards were prepared by mixing UF resin with graphene oxide. As a result, adding graphene oxide to UF resin improved IB (two times more than control), MOR, MOE and TS of the panels compared with the control and standard.

All particleboard panels made by mixing UF resin with the minimum content of graphene oxide conformed to the strength requirements of the European standard (P1, P2, P3). But the best results of mechanical and physical properties were obtained with 0,75 % GO mixed with UF resin.

By using graphene oxide, the shorter pressing time still provided benefits. Therefore, comparing two pressing time, 4 minutes was preferable than 5 minutes without having a negative effect on the mechanical and physical properties of the particleboards.

Authorship contributions

S. M. M.: Conceptualization/ Methodology/ Formal analysis/ Investigation/ Writing –original draft. L. J.: Conceptualization/ Methodology/ Validation/ Writing – review & editing/ Supervision/ Project administration.

F. F.: Methodology/ Formal analysis. A. A.: Conceptualization/ Methodology/ Formal analysis.

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