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Eco-friendly Mimosa Tannin Adhesive System for Bagasse Particleboard Fabrication

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THE main goal of this paper was to replace most paraformaldehyde (PF) by mimosa tannin based resins as a binding agent in particleboard manufacture. Physical and mechanical properties of the fabricated particleboards were studied to evaluate their quality. The physical properties involved thickness swelling and water absorption, whereas the mechanical properties involved internal bond strength, modulus of elasticity and modulus of rupture. The impacts of bagasse mesh size and polymethyl methacrylate (PMMA) as a coating material were also investigated. Particleboard resulted from mimosa tannin coated with PMMA had superior properties than the particleboard prepared from uncoated particles. The particleboard prepared from bagasse through mesh size 4 had much better qualities than the particleboard prepared from bagasse through mesh size 3 for all physical and mechanical properties at all tannin resin ratios content studied. The results also confirmed that, mimosa tannin could be used up to 12.5% resin without deteriorating the physical and mechanical properties of boards. In general, mimosa tannin–PF resin can be effectively used as adhesives for particleboard with reducing about 63% of PF consumption.

Keywords: Mimosa tannin, Paraformaldehyde, Adhesive, Bagasse, Particleboard, Characterization.

Introduction

The benefits of using lignocellulosic fibers in the composites have been greater than before owing to their comparative cheapness, their aptitude to recycle and their strengthening value [1,2]. Also, using the natural fibers in composites minimizes the waste disposal problem and reduces wood consumption consequently lowering the production costs. The natural fibers strengthened polymer composites are ecologically friendly materials having excellent specific strength, greater modulus, reduced density, low production cost, high corrosion resistance, high creep resistance, high toughness and biodegradability [**3**]. Formaldehyde-based resins including urea formaldehyde; phenol formaldehyde and melamine–urea–formaldehyde are the three most ordinarily resins used as binders in wood composites manufacturing. These adhesives are considered the best important variety of the characterized aminoplastic resins [4,5].

Recently, formaldehyde has been characterized as a human carcinogenic material and its use could be limited in the future [6]. Several trials have been done in order to decrease or substitute formaldehyde emissions in adhesive formulations by new natural adhesives materials [7-10]. Recently, vegetal tannin resins, lignin adhesives, and soy protein are the main natural resins used in wood board manufacture. There is a growing interest in tannin based resins. Tannins are naturally polyphenolic compounds existing in wood barks [11-13]. Tannins are natural hydrophilic complexing agents, which have been a subject

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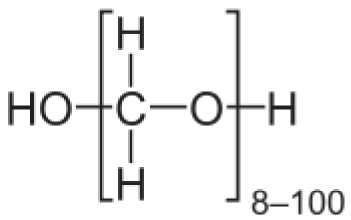
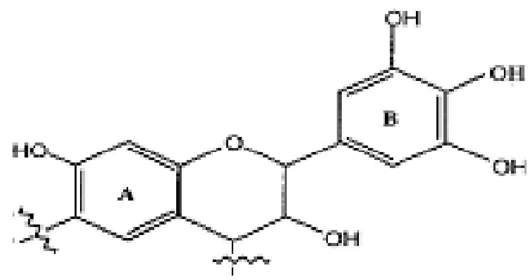
of wide research leading to the development of a varied range of industrial applications. Wattle bark tannins (*Acacia mearnsii*) adhesives were used safely to bind hardwood species [14,15].

Formaldehyde-based adhesives as phenol-formaldehyde and urea-formaldehyde resins are synthetically made from non-renewable resources such as petrol and natural gas. With declining petroleum stock and unbalanced fossil fuel charges, uncertainty concerning the upcoming price and accessibility of synthetic adhesives exist [16]. Numerous studies have been accepted to replace or substitute formaldehyde contents in adhesive formulations [17-19]. Many studies have confirmed that the phenolic nature of tannin makes it suitable for the production of polymeric resins and adhesives. About (30-50)% W/W tannin replacement of the amino and phenolic adhesives have been formulated into wood adhesives [20]. Saayman, [21] reported that tannin (Wattle tannin) utility in formaldehyde-based resins for wood adhesives requiring up to 30% of paraformaldehyde fortification to obtain a fully water–resistant bond. Wood adhesives from condensed flavonoid tannins have been established in other studies [22]**.** The reaction of tannin (Wattle tannin) with formaldehyde to synthesis polymeric resins was investigated [23,24]. Mimosa tannin is alternative renewable adhesive material which may be used for substituting petroleum-derivative phenolic compounds. Mimosa tannins (*Acacia mearnsii*) have higher reactivity with formaldehyde and have been used as a fortification to formaldehyde-based resins in order to produce particleboard and plywood [25]. The flavonoid units in mimosa tannins and paraformaldehyde are presented in Fig. 1 [25].



Paraformaldehyde



Mimosa tannin (Acacia mearnsii) flavonoids

Fig. 1. The chemical structure of mimosa tannin (Acacia mearnsii) flavonoids and paraformaldehyde.

The target of this research was to examine the replacement of most paraformaldehyde by mimosa tannin based resins as an adhesive in particleboard manufacture. The quality of this binder was evaluated by studying their physical properties (water absorption and thickness swelling) and mechanical properties (modulus of rupture "MOR", modulus of elasticity "MOE", internal bond strength). The effect of mesh size and PMMA, as a coating material, were also investigated.

Experimental

Raw material

Bagasse fibers were obtained from Quena Mill for papermaking, Upper Egypt. The Chemical constituents of bagasse fibers were found to be (α-cellulose 42%, pentosan 30%, lignin 22%, and ash 1.8%). The tannin extract used for the preparation of the resin was commercial mimosa tannin extract (non sulfited, hot water extracted) from Silva Team (San Michele Mondovi, Piedmont, Italy). Polymethyl methacrylate (PMMA) (granules of Mw~120,000, Sigma-Aldrich), paraformaldehyde and other chemicals were of pure analytical grades chemical and used without farther purification as received.

Methods

Preparation of tannin adhesives

To prepare a tannin adhesive, 10g mimosa tannin /111mL H_2O (pH≈7 was adjusted by using NaOH) was blended with 6 g paraformaldehyde under stirring.

Particleboard preparation

Particleboards of dimension 200 mm x 200 mm x 15 mm were prepared. The total tannin adhesive solid by weight were 5, 7.5, 10, and 12.5 % based on bagasse raw material. Hot pressing was carried out at 100 kg/cm^2 pressure and 190 °C, for 3.5 minutes press time, 50 kg/cm^2 for 2 minutes and 25 kg/cm^2 for 2 minutes. Particles were dried to approximately 3 % moisture content prior to application of resin. The particleboard density was 657 kg/m^3 which could be considered as a MDF.

Preparation of PMMA coated particleboard

PMMA was dissolved in toluene under mechanical stirring for 5 hours, forming a 10 wt. (%) solution. The prepared bagasse-tannin particleboards were immersed in the coating slurry for 1 min. Finally, the coated particleboards were dried at room temperature in air for 12 hours.

Composite property testing

Three replicates were tested for each property under each formulation. Thickness was measured using a dial micrometer.

Specimens for testing

After hot pressing, boards were conditioned at 20°C and 65% relative humidity for 72 h and then trimmed and cut into specimens of specific dimensions suitable to various tests. The specimens were conditioned again at a temperature of 20°C and 65% relative humidity for 72 h. Testing of specimens was carried out according to EN standards.

Water absorption and swelling measurements

Water absorption (WA) and thickness swelling (TS) were determined according to EN (317–1993) [26]. For water absorption, a small sample of the composite was immersed in water at room temperature for different times. After that, the sample was dried between two sheets of filter paper and then the weights as well as the dimension of the samples were measured. The percentage weight gain (PWG) was calculated as:

$$PWG (\%)=(W_f-W_o)/W_o\times100$$

Where W_o is the weight of the sample before the impregnation and W_f is the weight of the sample after the impregnation. The percentage of swellability was calculated as:

$$\text{Swellability } \% = (x-y)/y\times100$$

Where x is the volume of the sample after the impregnation and y is the volume of the sample before the impregnation.

Mechanical properties of the prepared composites

For mechanical properties, modulus of rupture (MOR) and modulus of elasticity (MOE) were measured according to EN 310, 1993 [27], and internal bond (IB) was measured according to EN 319, 1993 [28], by using LLOYD INSTRUMENTS LR 10K universal testing machine. Five specimens from each sample were tested, and results were averaged.

Scanning electron microscope (SEM)

Fracture surfaces of composite specimens were investigated via SEM with a JEOL JXA-840A electron probe micro-analyzer (Tokyo, Japan). The samples were coated with a thin layer of gold before SEM with an S1SoA Edward, sputter coater (Crawley, UK).

Results and Discussion

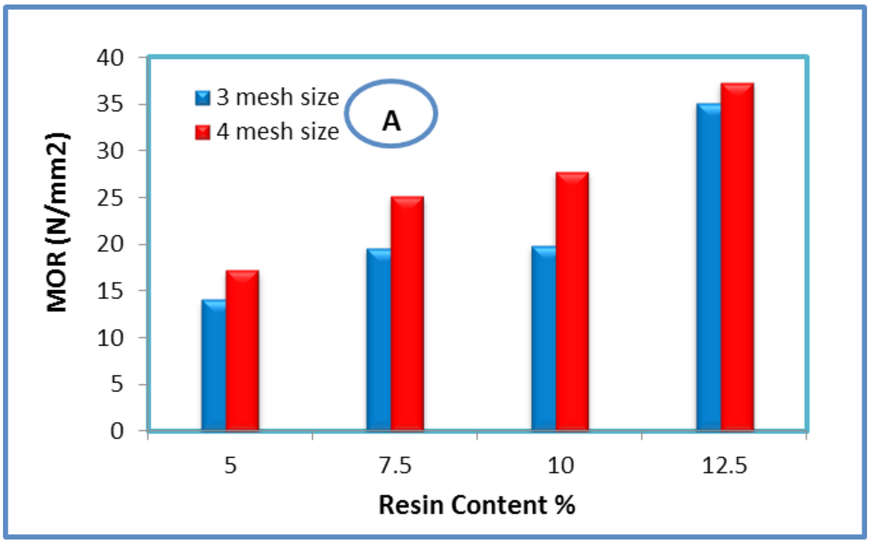
Tannin -bonded bagasse particleboard

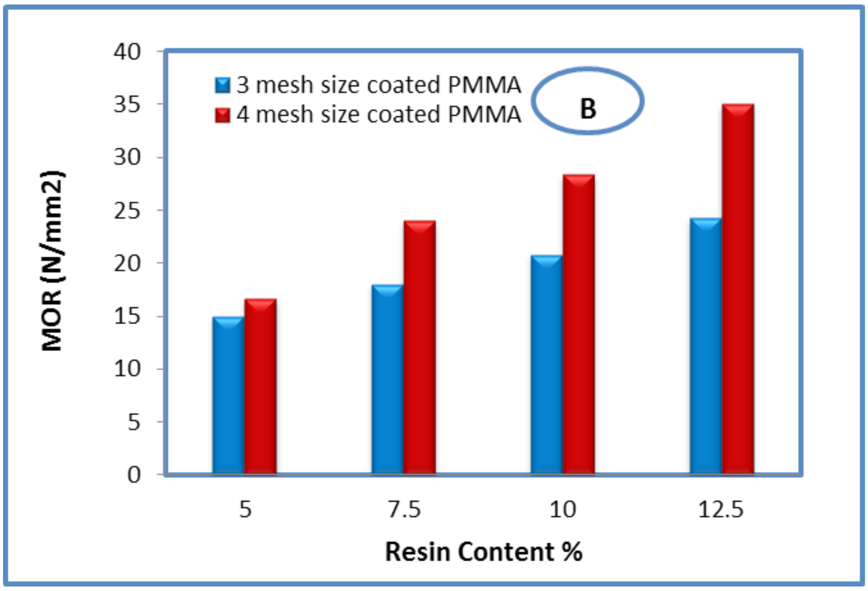
The bond quality is mainly affected by the pressing conditions and the curing of the resin. The pressing conditions must be adjusted to get boards with optimum mechanical and physical properties. After adjustment of these conditions, a comparison was achieved on a particle of mesh size of 3 and 4 at various tannin contents (5, 7.5, 10, and 12.5%).

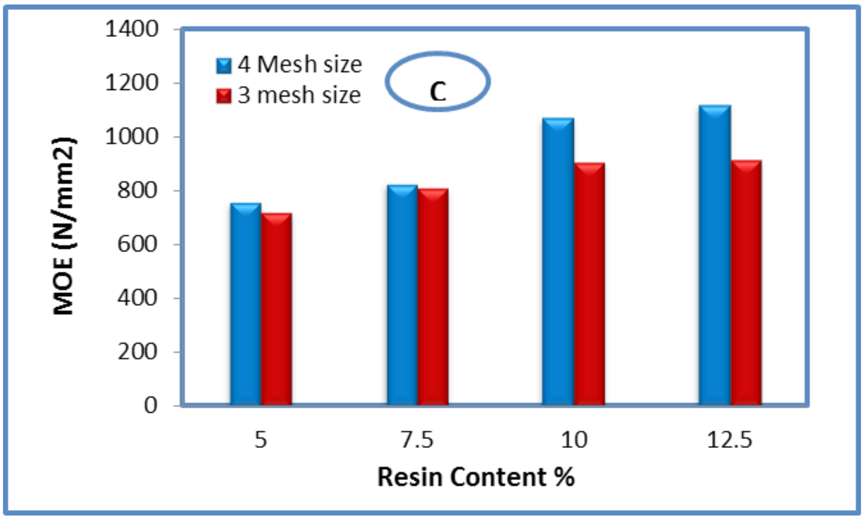
Mechanical properties

The dependence of mechanical properties of the tannin-bagasse particleboards (coated/uncoated with PMMA) made by using two types of bagasse particle mesh size (3 and 4) with different tannin contents were investigated and illustrated in Fig. 2.

From Fig. 2, it is clear that increasing tannin content caused significant improvement in the mechanical properties for coated and uncoated particleboard. MOR values for uncoated particleboard ranged from 14.1 to 35.3 and from 17.2 to 37.3 N/mm^2 for particle mesh size 3 and 4 respectively, and for coated PMME particleboard range from 15 to 24 and from 16.6 to 35.5 N/mm^2 for particle mesh size 3 and 4 respectively. MOE values for uncoated particleboard range from 760 to 1120 and from 720 to 915 N/mm^2 for particle mesh size 3 and 4 respectively, the MOE values of coated particleboard range from 950 to 1350 and from 856 to 1200 N/mm^2 for particle mesh size 3 and 4 respectively. The IB values of uncoated particleboard range from 0.23 to 0.39 and from 0.3 to 0.6 N/mm^2 for particle mesh size 3 and 4 respectively, the IB values for coated particleboard range from 0.13 to 0.23 and from 0.16 to 0.28 N/mm^2 for particle mesh size 3 and 4 respectively. It can be seen


MOR (N/mm2)
40
35
30
25
20
15
10
5
0
3 mesh size
4 mesh size
A
5
7.5
10
12.5
Resin Content %


MOR (N/mm2)
40
35
30
25
20
15
10
5
0
3 mesh size coated PMMA
4 mesh size coated PMMA
B
5
7.5
10
12.5
Resin Content %


MOE (N/mm2)
1400
1200
1000
800
600
400
200
0
4 Mesh size
3 mesh size
C
5
7.5
10
12.5
Resin Content %

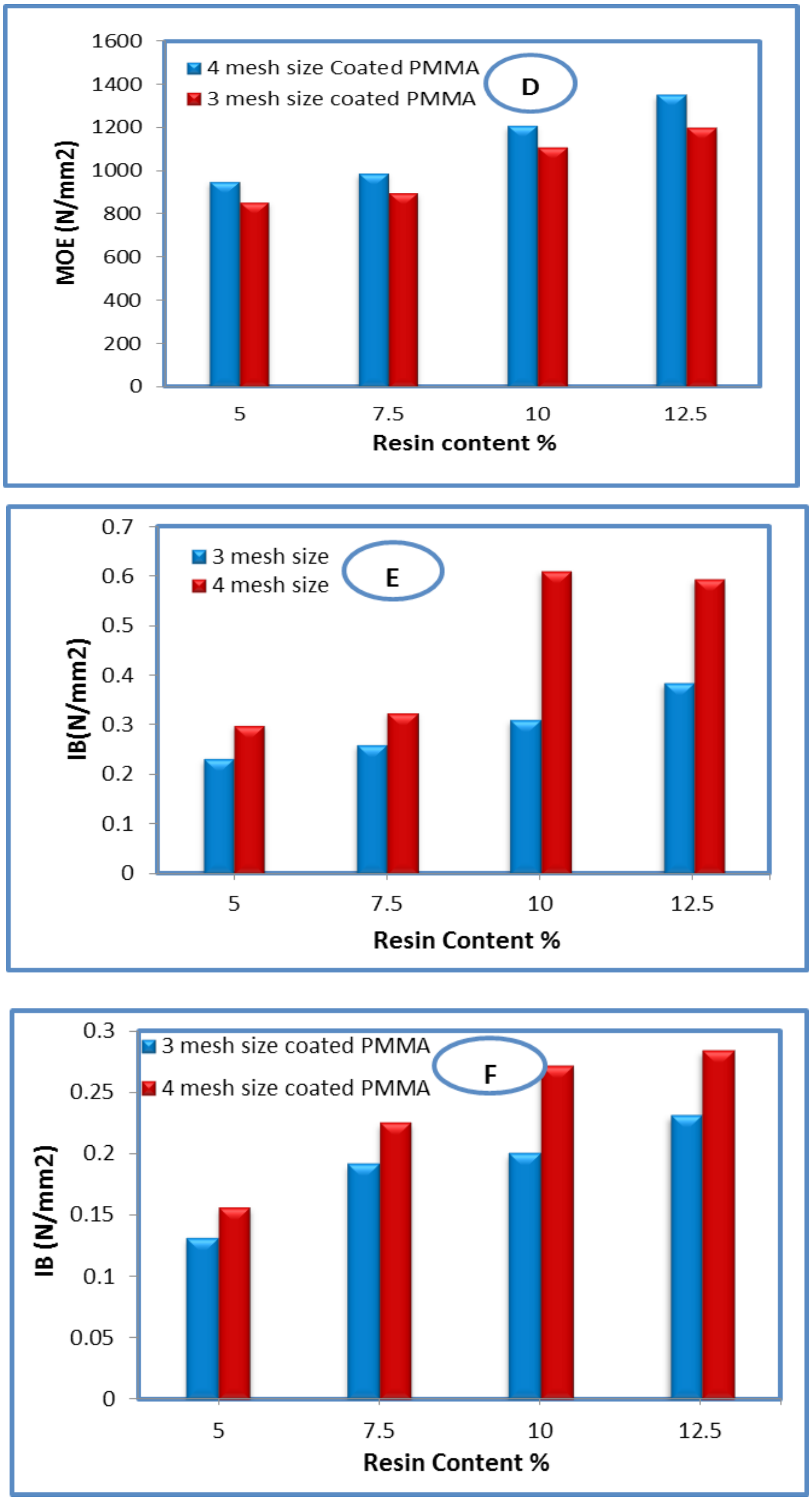


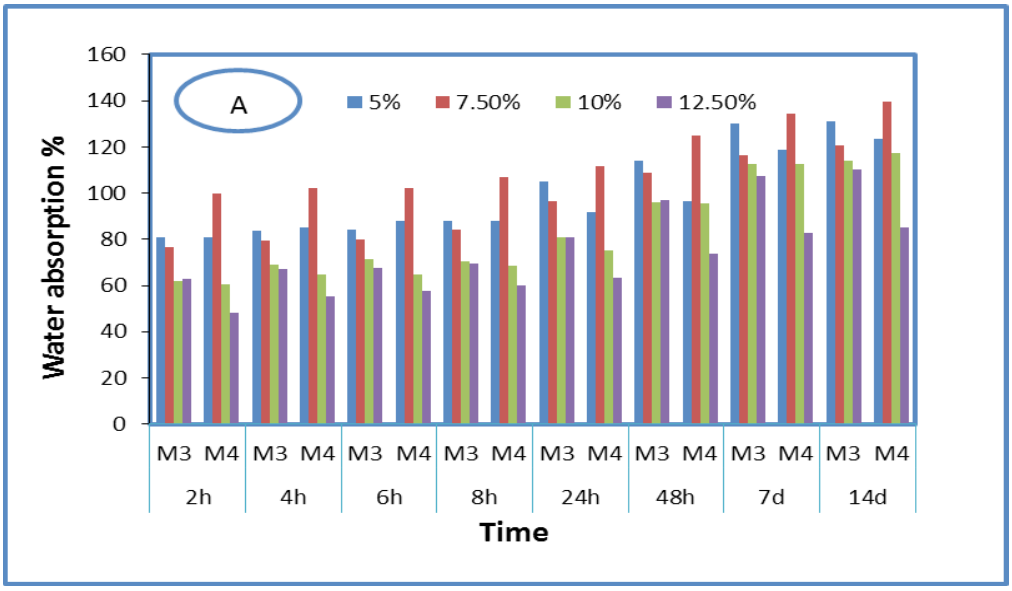
Fig. 2. Effect of resin content on the mechanical properties of; A) uncoated board (MOR), B) coated board with PMMA (MOR), C) uncoated board (MOE), D) coated board with PMMA (MOE), E) uncoated board (IB), F) coated board with PMMA (IB).

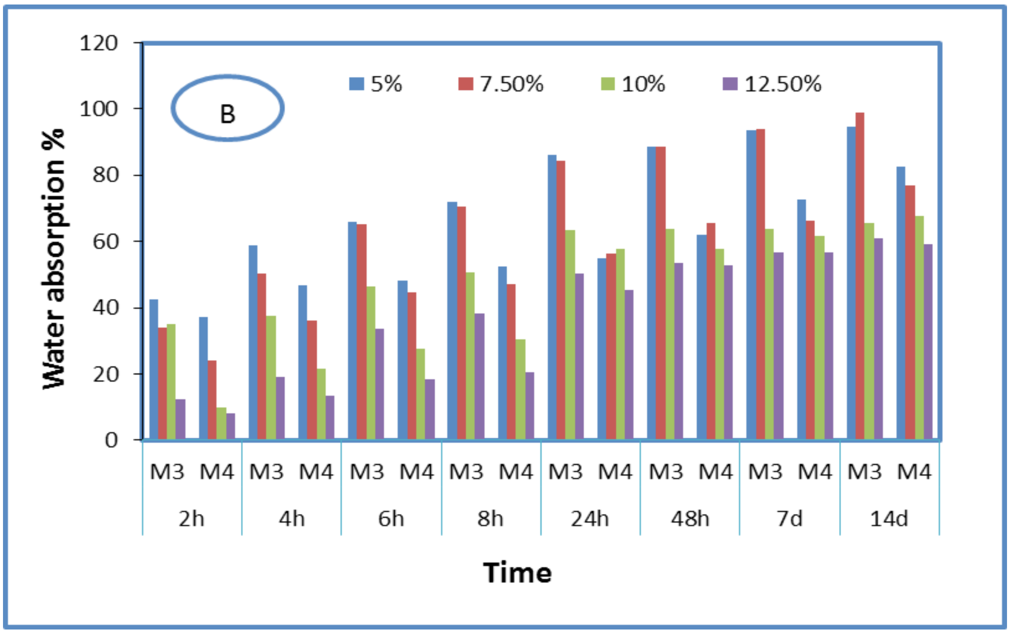
from the figure that the MOR, MOE and IB differed significantly with the particle mesh size and obviously improved with increasing resin contents. This improvement could be described by the distribution of the adhesive surrounded by the particles over wide particle surface as well as in the curing of the adhesive [29]. Furthermore, PMMA coating have no serious effect in MOR properties, while MOE are slightly improved and IB decreased as a result of PMMA coating.

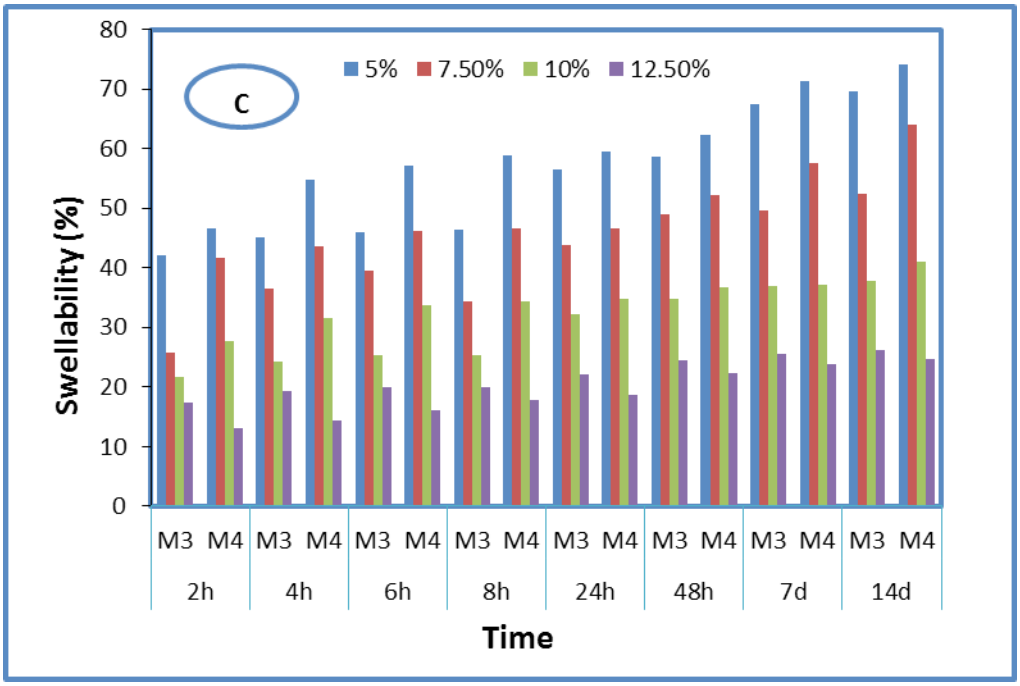
Physical properties of tannin-bagasse particleboards

Figure 3 illustrates the dependence of water absorption (WA) and thickness swelling (TS) of prepared tannin-bagasse particleboards (uncoated/coated with PMMA) using different tannin concentration.

With respect to physical properties, it was observed that both water absorption and thickness swelling were increased as the immersion time of boards in water increased. The expected results of decreasing water absorption and thickness swelling by increase of boards' resin content was evidently observed, especially at short immersion time. The particleboard prepared from bagasse mesh size 4, in general, had superior physical properties than the particleboard prepared from bagasse mesh size 3 at all tannin resin ratios







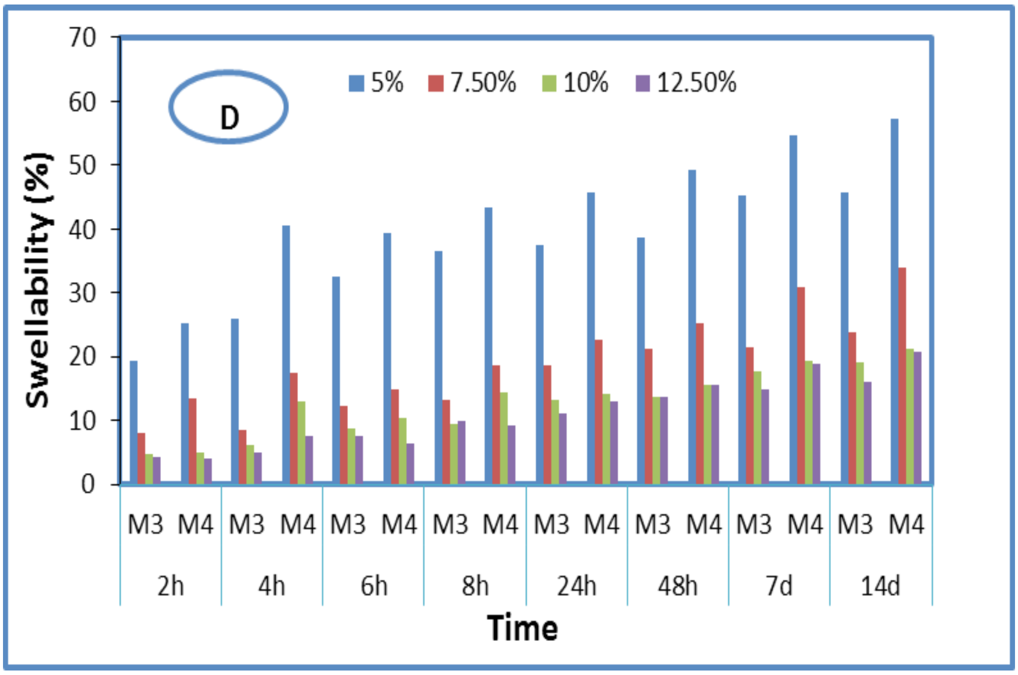


Fig. 3. Water absorption (WA) and thickness swelling (TS) of; A) uncoated board (WA), B) coated board with PMMA (WA), C) uncoated board (TS), and D) coated board with PMMA (TS).

content studied. Superior physical properties were attained by coating boards with PMMA, an obvious decrease in water absorption and considerable reduction in thickness swelling were recorded. This may be attributed to the covering layer of PMMA on the bagasse tannin board as an insulating layer for water absorption, where the ratio of water absorption sharply decreased by adding PMMA as presented in Fig.3.

Scanning Electron microscope (SEM)

In the current article, prepared bagasse tannin particleboards were investigated by SEM as showed in Fig. 4(A-D). The figures show that

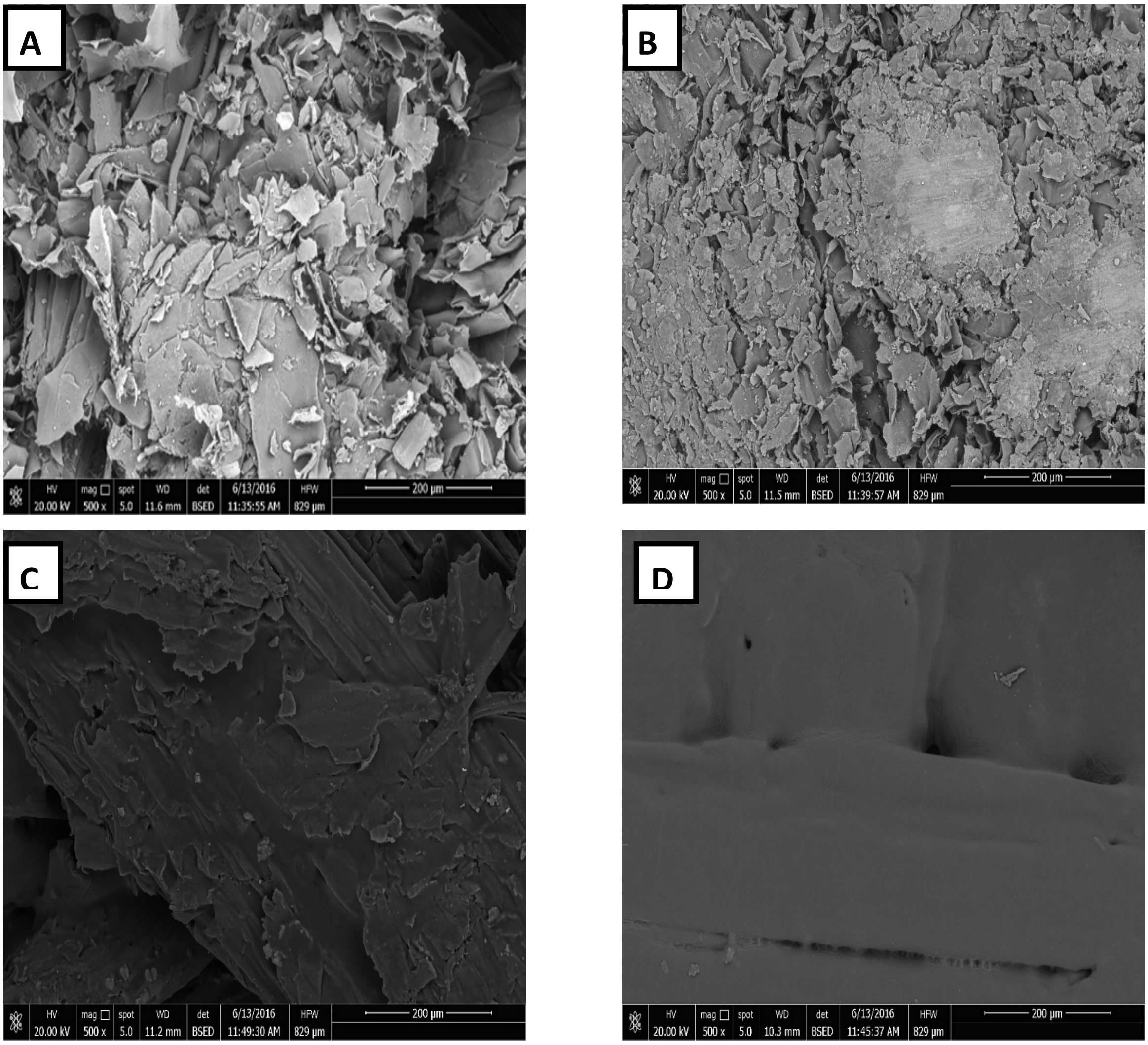


Fig. 4. SEM photographs of tannin particleboard a) mesh size 3 PMMA uncoated b) mesh size 4 PMMA uncoated C) mesh size 3 coated with PMMA and d) mesh size 4 coated with PMMA.

the fibers are spongy in shape, which permits greater surface area in contact with the resin used; consequently more bonding between the fibers and the resin is expected. Smoother surfaces were observed for boards have mesh size 4 (Fig. 4 B&D) compared with boards have mesh size 3 (Fig. 4 A&C), also the coated boards with PMMA (Fig. 4 C&D) were homogeneous and glossy compared with the uncoated boards (Fig. 4 A&B).

Effect of particle size on particleboard properties

The impact of particle size on particleboard properties was tested by using two different mesh sizes, 3 and 4. Figure 2 illustrates the influence of particle size on the mechanical properties of the resulting boards. It is obvious from the figure that shorter fibers (mesh 4) offer higher internal bond strength and modulus of rupture. Also, modulus of elasticity is somewhat higher for boards made with fiber of mesh size 4. Furthermore, physical properties are enhanced when using bagasse fibers has mesh size 4, compared with those of mesh 3 as displayed in Fig 3. The use of the low size of bagasse fibers in the preparation of the particleboards led to better properties than the use of larger size. The shorter the fibers size of the bagasse in the particleboard, the greater the adsorption of the resin and the better encapsulation of the resin around the fibers. This may lead to the use of a lower amount of the resin and, of course, more powerful particleboards and better properties are obtained than using larger sizes. Therefore, the stiffness of the boards should be increased which will result in a higher modulus of elasticity.

It is a well-established that fiber length is a critical parameter in evaluating the properties of cellulose fiber-particleboard materials [30]. The short fibers, i.e. fibers with critical length, prevent entanglement while fiber characteristics are maintained. Moreover, such fiber length provides better green strength, high dimensional stability, resistance to break during fabrication in addition to homogeneous mixing with resins, etc. [31].

Conclusions

The present investigation on the using of mimosa tannin resin with a minimized PF level to prepare mimosa tannin–PF resin shows that particleboard with acceptable mechanical and physical properties could be obtained by adding up to 12.5% of mimosa tannin resin, with the additional advantage of more ecofriendly products. Mimosa tannin–PF resin can be used successfully as a wood adhesive for constructing particleboard with about 63% reduction of PF consumption. Research is in progress to replace synthetic wood adhesive by employing other natural renewable resource.

Acknowledgement

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نظام لاصق صديقا للبيئة من الميموزا تانين لتصنيع الواح حبيبة من قصب السكر

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قسم السليلوز والورق - المركز القومي للبحوث - 33 شارع البحوث (ش التحرير السابق) - الدقي -
ص.ب. 12226 - الجيزة - مصر.

كان الهدف الرئيسي من هذا العمل هو تقييم استبدال نسب مختلفة من مادة البارافورمالدهيد باستخدام راتنج الميموزا تانين كمادة لاصقة في تصنيع الالواح الخشبية الحبيبية. ومن أجل تقييم جودة هذه المادة في الالواح الناتجة، تمت دراسة الخواص الفيزيائية والميكانيكية للألواح الناتجة. وقد شملت الخواص الفيزيائية كلا من امتصاص الماء ونسبة الزيادة في السماكة، في حين تضمنت الخواص الميكانيكية قوة الرابطة الداخلية، ومعامل التمزق ومعامل المرونة. كما تمت دراسة تأثير حجم شباك قصب السكر وبولي ميثيل ميثاكريلات كمواد طلاء. أوضحت النتائج أن الالواح الحبيبة الناتجة من الميموزا التانين والمغلفة بواسطة مادة البولي ميثيل ميثاكريلات كانت لها خوص متفوقة من الألواح المصنوعة من جسيمات غير مغلفة. علاوة على ذالك وجد ان الالواح المصنوعة من شبكة مصاصة القصب ذات حجم ٤ يتمتع بصفات أفضل بكثير من الالواح المصنوعة من حجم فتحات الشباك ذات حجم ٣ وذالك لجميع الخواص الفيزيائية والميكانيكية في كل نسب محتوى التانين الراتنجية التي تمت دراستها. واكدت الدراسة ايضا انه من الممكن استخدام مادة الميموزا التانين، إلى نسبة تصل لحوالى ١٢,٥ ٪ من الراتنج المستخدم دون تدهور في الخواص الفيزيائية والميكانيكية للألواح الناتجة. وبشكل عام، يمكن استخدام راتنج الميموزا تانين مع مادة البارافورمالدهيد بنجاح كمواد لاصقة للخشب لإنتاج لوح الخشب الحبيبي مع تقليل حوالي نسبة ٩٤٪ من استهلاك مادة البارافورمالدهيد المستخدمة في هذه الصناعة.