PEER-REVIEWED ARTICLE *Lignocellulose*

THE EFFECT OF FIBER MOISTURE AND DRYING TEMPERATURE ON HARDWOOD FIBER PHYSICAL CHEMISTRY AND STRENGTH OF MEDIUM DENSITY FIBERBOARD

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The effect of hardwood thermo-mechanical fibers drying condition (drying temperature; 110, 130, and 150°C and fiber moisture content; 50%, 60% and 70%) on fiber physical chemistry and bond formation was studied. The results revealed that at higher drying temperature, the pH of dried fibers increased marginally, but fiber moisture content was more influential and at higher moisture content especially when higher drying temperature was applied, pH was higher. The pH varied between 4.71 and 6.11. Acid buffering capacity of fibers containing lower moisture and dried at lower temperature was more than acid buffering capacity of fibers from other treatments. The amount of acid buffering capacity varied between 0.67 and 2.05 ml 1N NaOHg-1 of fibers. The effect of drying condition on fiber wettability revealed that milder conditions generated better wettability, as indicated by higher water rise in capillary tube. The flexural strength and bond development between fibers dried at treatments with applying milder condition (110ºC drying temperature) were superior to identical properties from other treatments and were best correlated with wettability of fibers.

Keywords: *Buffering capacity, flexural strength, internal bonding, pH, wettability*

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INTRODUCTION

Medium density fiberboard (MDF), which was developed and introduced during 1970's has been among the most successful wood based panel board product, thanks to its unique specification and performance including strength, homogenization and machining behavior (Halversson et al. 2008). Such unique specifications opened its way into various applications and generated interest among different consumer sectors especially cabinet and home furniture. MDF frequently replaces solid wood, plywood as well as particleboard. During the past four decades, the MDF industry has expanded rapidly due to development of reactive resins and process modifications which helped both strength enhancement and cost reduction. Consequently, MDF producers have been able to fulfill the needs of consumers to their satisfaction and expand the market. As a result, world MDF production was grown to almost 55.8 million m3 in 2009 (FAO 2010).

MDF expansion necessitated extensive research and development by those involved in this sector. Current research and development activities are following two general paths; diversifying the raw material supply and enhancing the product quality.

Raw material (wood and binder) and energy have been the major determinants in MDF production, especially the availability of wood. However, during the course of MDF production expansion and the implementation of new production facilities around the globe, and declining the availability of wood, a widening in-balance between supply and demand for wood has developed, which imposes tough condition on this sector. In addition, competition between wood as an industrial feedstock and as an energy source is increasing, which forces the whole wood chain to reorganize the raw material consumption and cascade use of wood and recovery of waste wood products (Roffael et al. 2010) and causes uncommon and unconventional raw material to be used. Fast growth, small diameter poplar and eucalyptus wood were the first candidates (Roffaelet al. 1992; Krzysik et al. 1999). Bamboo was investigated, and promising results were reached. Thus, forest-deficient countries like Iran and Egypt had implemented MDF projects based on bagasse (Jahan Latibari and Roohnia 2010). Fortunately, in contrast to previous attempts by China, Thailand and Pakistan, these projects had reached successful production. Attempts to utilize the potential of agro-based raw material in panel board production such as MDF are still continuing, and an excellent wealth of research and development reports has become available (Akgul and Toslughu 2008; Copur et al. 2008; Lee et al. 2006; Ye et al. 2007; Halvarsson et al. 2010; Pan et al. 2010; Ciannamea et al. 2010).

Enhancing the production technology has been another path in process optimization to improve bond formation potential through in depth understanding of process and material variables. A recent research trend has been aimed at improving the characteristics of MDF. Whilst this can be achieved using high quality wood and binders as well as implementing specific measures during pressing, reducing the density of MDF boards has been an alternative both for cost saving and product improvement (Barbu and Resch 2000). Other factors that influence the strength of the MDF as a composite material are the inherent strength of its components; fibers and the bond strength between fibers and binder. (Halvarsson et al. 2008). Therefore, alternative defibration processes to produce stronger fibers have been investigated, and it has been discovered that chemi-thermo-mechanical pulp fibers produce stronger MDF and higher steaming temperature in thermo-mechanical pulping imposes negative impact on bond strength (Roffael et al. 2000). Apart from the typical methods involving both physical and chemical (pH and buffering capacity) parameters, the wettabilty of the fibers has be acknowledged as a helpful index to understand bond formation and enhancement (Kowaluk et al. 2008; Jarusombati et al. 2010; Roffael et al. 2010).

The objective behind this research was focused on determination of the influence of two important processing variables; fiber moisture content after refining and drying temperature on chemical behavior of dried fibers (pH, and buffering capacity), fiber wettability and the strength of manufactured MDF.

EXPERIMENTAL

Raw material:

- TMP fibers produced from Northern Iran forest hardwood species were collected from the outlet of start-up cyclone at a MDF production mill. Wet fibers were transferred to Wood and Paper Research Laboratory, Islamic Azad University, Karaj Branch. Wet fibers were

dried at ambient temperature to reach about 20% moisture content (wet basis), and then packed into polyethylene bags until used. The fiber composition of hardwood fibers was measured as followed:

Sieve opening (mesh)	R5 (4mm)	R10 (2mm)	R18 (1mm)	R40 (0.415mm)	R50 (0.300mm)	R70 (0.212mm)	P70 (0.212mm)
Fraction (%)	3.30	12.67	22.27	20.25	12.23	12.27	26.50

- Urea-formaldehyde resin at 63% solid content, and specific gravity, viscosity, gelation time and pH of 1.26 gcm^{-3}, 69 seconds, 81 seconds and 8.15 respectively was supplied by a local resin manufacturing plant (Fars Chemical Co.).

-Reagent grade ammonium chloride (20% solution) was used as hardener.

Fiber drying

Fibers were rewetted to target moisture content (50%, 60% and 70%, wet basis) by spraying the pre-determine volume of water prior to final drying. A laboratory rotating drum dryer (Faravari Ghomes Co.) equipped with electrical heating elements was used to dry the fibers to reach the final moisture content of 3% (dry basis). Temperature was controlled using digital thermostat. Drying temperatures were adjusted at 110, 130 and 150ºC. Dried fibers were stored in polyethylene bags until used.

Acidity and wettability measurements:

The pH and both acid and alkaline buffering capacity of fibers were measured following the procedure introduced by Johns and Niazi (1980). Dried fibers were extracted using de-ionized water without any further grinding. The water extract was used for pH and buffering capacity measurements.

Wettability of fibers was measured using a column wicking technique (Shen et al. 2004). The glass measuring tube with the inner diameter of 4 millimeter and the length of about 25 centimeters was used. In any measurement, a pre-weighted amount of 1.5 grams of dried fibers were uniformly packed into the length of 18 centimeters in the tube with the help of a thin rod. After packing, the column was vertically placed in a beaker containing distilled water at room temperature. The penetration height (water rise) was measured when it reached the equilibrium height. Each set of measurements were conducted simultaneously so the effects of ambient temperature and relative humidity at different measuring times were eliminated (Fig.1).

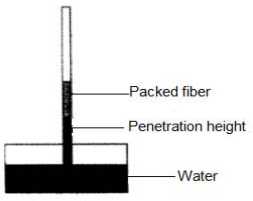

Fig. 1. Illustration of wicking column experiment set up

Board making and evaluation

Fibers were blended with 10 % resin (dry basis) and 2% hardener (based on dry weight of resin) utilizing rotary drum blender equipped with spray nozzle. A pre-weighted amount of

resinated fibers was then hand-formed, simulating industrial forming, using a 400x400 mm wooden box. Board target density and thickness were selected at 0.75 gcm^{-3} and 12 millimeters, respectively. Mats were cold per-pressed and then hot pressed to 12 millimeter final thickness, using a laboratory hydraulic press (Pishroo Machine Manufacturing Co.) applying 40 bar specific pressure and five millimeters per second closing speed. Press temperature was set at 175°C and four minutes press time was applied. At the end of press cycle, boards were discharged and cooled at room temperature. Three boards for each combination of variables and total of 27 boards were produced.

Test sample were prepared from each board according to EN325-1 standard. Modulus of rupture (MOR) and modulus of elasticity (MOE) were measured according to EN310/1996, and Internal bonding (IB), EN319/1996 standards. Universal testing Machine (Payam Goster Hoshmand PT500) was used for strength measurement.

Statistical analysis

Factorial experimental design in a completely randomized was used for statistical analysis of the generated data. In case, significant difference at either 99% or 95% level was observed, then DMRT mean separation of averages was applied.

RESULTS AND DISCUSSION

The pH and both acid and alkaline buffering capacities of hardwood fibers dried at different moisture content together with drying temperatures and fiber moisture content are summarized in Table 1. The results indicate that drying temperature influences the pH of dried fibers significantly (Table 2). The moisture content of the fibers prior to drying was also influential on pH. Acid buffering capacity which is an important variable on urea formaldehyde resin curing was reduced as the fiber moisture content increased.

Table 1. pH, buffering capacity and wettability of hardwood MDF fibers dried at different temperatures and moisture contents

Trial No.	Drying Temperature (°C)	Moisture Content (%)	pH	Acid buffering (ml 1N $NaOHg^{-1}$)	Alkaline buffering (ml 1N $H_2SO_4g^{-1}$)	Penetration height (cm)
T1	110	50	5.06^{ab*}	2.3^{e}	0.12^{ab}	15.3^{d}
T2	110	60	6.16^{c}	1.60^{d}	0.14^{abc}	11.3^{ab}
T3	110	70	5.04^{ab}	1.85^{de}	0.11^{a}	13.5^{c}
T4	130	50	5.04^{ab}	1.34^{bcd}	0.20^{d}	12.5^{bc}
T5	130	60	5.31^{b}	0.67^{a}	0.14^{abc}	10.8^{a}
T6	130	70	5.30^{b}	0.85^{abc}	0.13^{abc}	13.3^{c}
T7	150	50	4.71^{a}	1.25^{abcd}	0.17^{bcd}	13.6^{c}
T8	150	60	5.05^{an}	1.40^{cd}	0.16^{abcd}	14.1^{cd}
T9	150	70	6.11^{c}	0.75^{ab}	0.19^{cd}	15.6^{d}

* Superscript case letters indicates the DMRT mean separation of averages

Wettability of the dried fibers is also summarized in Table 1 and the statistical analysis of the data is provided in table 2, indicating that the effect of both variables on wettability of fiber is statistically significant at 99% level. Higher moisture content and higher drying temperature produced higher wetting in fibers indicated by higher water rise in capillary tube.

The bending strength (MOR) and modulus of elasticity (MOE) as well as internal

bonding (IB) of the MDF boards produced from dried fibers are shown in figures 2-4. Each value in figures 2-4 is the average of 12 measurements. In order to reveal the statistical differences among the measured values, the significance levels are shown in Table 2. In case a statistically significant difference between averages was observed, Duncan multiple ranges grouping of the averages are shown in each figure using lower case letters.

Table 2. Analysis of variance of the results of measurement

Source	Degree of freedom	F-values						
		Penetration height	pH	Acid Buffering Capacity	Alkaline Buffering Capacity	MOR	MOE	IB
Drying temperature (DT)	2	14.33**	4.02*	22.16**	7.35**	13.67**	84.5**	7.69**
Fiber moistures content (MC)	2	13.48**	19.51**	5.54**	1.37ns	18.22**	37.9**	4.63**
DT*MC	4	5.67**	22.52**	2.31ns	3.17*	12.20**	50.9**	3.78**

** Significant at 99% * Significant at 95% ns: non-significant

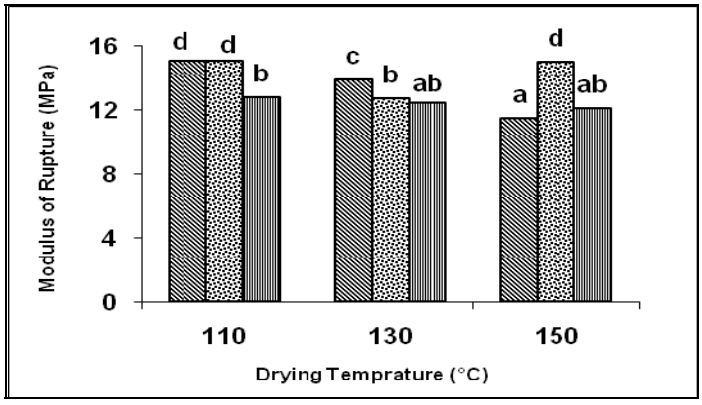

Fig. 2. The influence of drying condition on modulus of rupture of the MDF boards produced from hardwood fibers (Fiber moisture content; ▨ 50%, ▩ 60%, ▥ 70%)

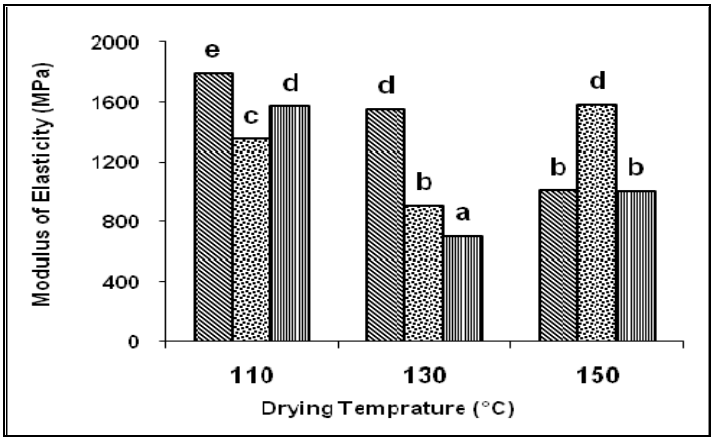

Fig. 3. The influence of drying condition on modulus of elasticity of the MDF boards produced from hardwood fibers (Fiber moisture content; ▨ 50%, ▩ 60%, ▥ 70%)

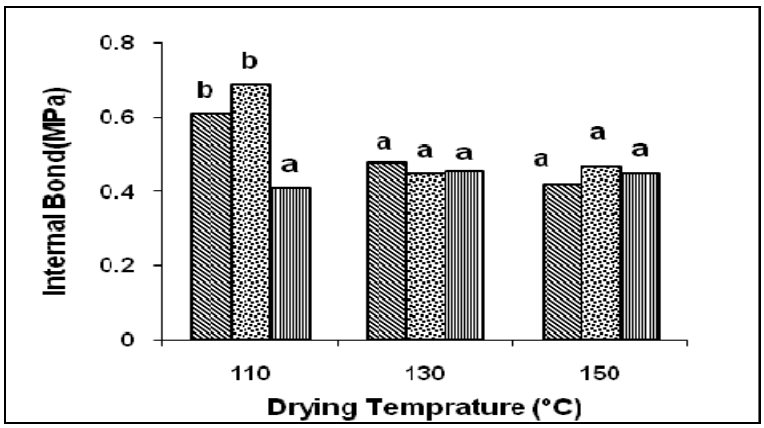

Fig. 4. The influence of drying condition on internal bond of the MDF boards produced from hardwood fibers (Fiber moisture content; ▨ 50%, ▩ 60%, ▥ 70%)

The curing reaction of pH-sensitive resins such as urea-formaldehyde initiates and propagates in acidic condition. Therefore, the knowledge of the pH and buffering capacity of lignocellulosic substrates is essential to the performance of urea-formaldehyde resin for proper bond development and durability. The gelation time of urea-formaldehyde resin is directly correlated to the pH and inversely depends on acid buffering capacity (Johns and Niazi 1980). The pH measurements of the dried MDF fibers showed that both fiber moisture content and drying temperature influences the pH of the dried fibers. The moisture content of the fibers prior to drying was influential on pH and at higher moisture content (T9), pH was higher probably due to deterioration of acidic compounds which possibly generated milder drying condition (lower drying temperature and higher fiber moisture). Acid buffering capacity, which is an important variable affecting urea-formaldehyde resin curing, was increased as the fiber moisture content increased, which indicates that partial hydrolysis of hemicelluloses occurs. The impact of drying temperature is also evident, and 130°C drying temperature generated lower amount of acidic compounds. One possible reason may be the fact that acidic components formed during the thermo mechanical fiber preparation and application of high temperature steaming (Pan, et al. 2010, Winandy and Smith, 2006) inhibits further hydrolysis during subsequent moist drying. The refined fibers pH is usually below 6 (in the range of 3.5-5.5) lower than agro-based material such as wheat straw (Halvarsson et al. 2010).

Even though the increase in either pH or acid buffering capacity show similar impact on the gelation times of urea-formaldehyde resins when in contact with fiber aqueous extract (Johns and Niazi 1980), but the pH of the water soluble extract from agro-based material does not reflect the best indication of the relative suitability of the material for urea-formaldehyde resin bonding system and buffering capacity of water soluble substances appeared to be the best indicator of the performance (Mclauchlin and Hague 1998). This may be reflected in wood based fibers as well.

Apart from pH and buffering capacity, wettability will provide the needed information for characterizing the bonding potential between fibers and resin (Kowaluk et al. 2008). Wettability is defined as the condition of the surface that determines how fast the resin will wet and spread on the surface. It is crucial for good bond formation (Jarusombuti et al. 2010). To determine the impact of fiber treatment (drying temperature and fiber moisture content prior to drying), the wettability of fibers was measured using the column wicking technique

(Table 1). It has been acknowledged that column wicking is influenced by different packing densities. However with respect to sample protection especially hygroscopic material such as lignocellulosic fibers, which absorbs moisture from the surrounding, column wicking provides advantages (Shen et al. 2004). Therefore to eliminate the adverse effect of packing density, glass tubes were filled and packed incrementally to be sure that packing was uniform.

The interaction of treatment variables (drying temperature and fiber moisture content) revealed interesting results. Lower drying temperature and lower moisture content produced higher wettability (T1), but as the drying temperature increased to 150°C, then higher moisture content generated better wettability (T9). Higher drying temperature lowers wettability of the fibers. Thermo hydrolysis of hemicelluloses during drying occurs and this phenomenon will decrease the hygroscopicity of fibers (Jarusombuti et al. 2010). Also, lignin which is present on thermo-mechanical fibers produced with applying high temperature during fiber production may be reactivate under higher drying temperature. Fibers dried at 150°C were darker than other treatments and the bulk density of these fibers were higher which exhibits fiber shrinking. However, as both drying temperature and moisture content increases simultaneously, the mild treatment condition is exerted on fibers which results in lower shrinkage of fibers and availability of higher number of hydroxyl groups for water absorption and consequently better wettability. This phenomenon generates hydrophilicity in dried fibers (Cetin et al. 1999).

The TMP fibers produced from hardwoods are usually very short, and therefore higher dosage of resin is applied. However to reflect the impact of fiber chemistry and wettability on fiber bonding and strength development, only 10% urea formaldehyde resin was used. Consequently the strength values, both internal bonding (Fig. 4) and flexural strength (Fig.2 and 3) were lower than usual values.

The strength of composite material such as MDF depends on inherent strength of its components and the strength of the bonds formed by adhesive (Halvarsson et al. 2008). For MDF, as composite material which is produced using fibers originated from lignocellulosic material, the bond failure should occur in fibers itself and if there is any interfacial failure, then the bond formation is not adequate and efficient. Therefore, stronger fibers produce stronger boards (Roffael et al. 2000). It has been acknowledged that elevated temperature and pressure applied in thermo mechanical MDF fiber preparation process improves the flexural strength and internal bonding of finished product due to stronger and more wettable fibers (Han et al. 2010). Since lower drying temperature and lower fiber moisture content imparts milder treatment on hardwood TMP fibers, then higher flexural strength (Fig. 2 and 3) and higher internal bonding (Fig. 4) is produced. This treatment condition also generates better wettability (Table 1) and consequently produces more efficient bond formation. Our results imply that wettability reflects better correlation with bond formation than acid buffering capacity.

In regard to lower than required values for either flexural strength and internal bonding, it should be mentioned that due to higher content of fine and short fiber fraction and dust in hardwood TMP fiber, because of the existence of parenchyma cell, ray cells and vessel elements (P50 fraction is almost 38.77%), not only the network strength reduces, but also the surface area of fibers increases and resin coverage is low (Lee et al. 2006; Mobarak et al. 1982; Hill and Wilson 1978; Xing et al. 2006).

CONCLUSION

With respect to this research, we need to mention that societies are facing shortage of wood supply, and the situation in countries located in forest deficient areas that are willing to operate board production facilities is severe. The situation will be harder in the era that forest rich countries are intending to use wood and other lignocellulosic residues for energy generation. This reflects the need to develop or modify procedures to enhance the bond formation between fibers and adhesive. As a result of better bond formation, the strength of MDF will be improved and efficient use of wood and other lignocellolusic material will be achieved.

Fiber moisture content and drying temperature have been considered as effective variable in MDF production. This study revealed that fibers containing 50% moisture content and dried applying 110 C generated suitable properties for board production. The pH, acid and alkaline buffering capacity and wettability of fibers dried at the above drying condition were measured as 5.06, 2.05 mL 1N $NaOHg^{-1}$, 0.12 mL 1N H_2SO_4, and 15.3 cm water rise. The strength properties of the MDF board produced using these fibers including MOR, MOE and IB were measured as 15.3MPa., 1798MPa. and 0.61MPa, respectively. Thickness swelling after 2 and 24 hour soaking in water were 16% and 23.6%.

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