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Particleboard is widely used for making bookshelf, furniture, cabinets and many other interior products. At present, particleboard is mainly produced with wood particles and urea-formaldehyde (UF) resins. The emission of carcinogenic formaldehyde in the production and use of particleboard has generated an urgent need for development of a formaldehyde-free wood adhesive for making particleboard. Formaldehyde is derived from natural gas whose reserve is finite. It is also desirable to develop a wood adhesive from renewable material for making particleboard. Soy flour is inexpensive, abundant, renewable and readily available. A formaldehyde-free wood adhesive consisting of soy flour (SF) and a curing agent (CA) CA1000 has been used for commercial production of interior plywood since 2004. However, this CA-soy adhesive has high viscosity and is difficult to be sprayed onto wood particles with a conventional spraying nozzle. This study developed a new method of using this CA-soy adhesive for making particleboard. This new method involved the coating of wood particles with a dilute soy slurry in water, the drying of the soy-coated wood particles, the spraying of the CA onto the dried soy-coated wood particles, the formation of a particleboard mat with the CA-soy-coated wood particles and the hot-pressing of the mat into particleboard. The high viscosity of the adhesive was no longer an issue with this new method. For investigation of the effectiveness of this new method, effects of particleboard density, adhesive usages for both core and face particles, the solids content of the soy slurry, hot-press time, hot-press temperature, the storage time of the soy-coated wood particles, and the soy/CA ratio on the internal bond strength (IB), the modulus of rupture (MOR), and the modulus of elasticity (MOE) of the resulting particleboard were investigated in detail. Results demonstrated that this new method had wide operational windows for making particleboard and allowed the strengths of particleboard bonded with this CA-soy adhesive to exceed the industry requirements of M-2 particleboard. The optimal conditions of using this method for making particleboard in terms of enhancing the IB, MOR and MOE were: 760 kg/m^3 of the particleboard density, 11 wt% resin usage for the core particles, 12 wt% resin usage for the face particles, 20 wt% solids content of the soy slurry, 180 °C of the hot-press temperature, 224 s of the hot-press time, 1:7 CA/SF weight ratio, and 36 h of the storage time for the wet soy-coated wood particles.

A New Method of Making Particleboard with a Formaldehyde-free Soy-based Adhesive

by

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I understand that my thesis will become part of the permanent collection of Oregon State University libraries. My signature below authorizes release of my thesis to any reader upon request.

Lapyote Prasittisopin, Author

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INTRODUCTION

Particleboard panels

General knowledge of particleboard panels

By a definition from the American Heritage Dictionary of the English Language, Fourth Edition (2003), particleboard means a wood-based panel that is produced from wood fragments such as chips or shavings, and adhesives via a hot-press process. More specifically, wood is first mechanically broken into small particles that are then blended with an appropriate amount of an adhesive. The adhesive-coated particles are formed into a loose mat that is further pressed at elevated temperature to form particleboard panels. The major difference between particleboard and other wood-based panel products such as plywood, oriented strandboard (OSB), fiberboard, and hardboard is the size of wood furnish.

In general, there are four types of particleboard panels: random (no distinct layers), graduated (gradual transition between layers), three-layer (finer particles for faces and coarser for cores), and five- or more layer particleboards (finer for faces, slender and flat for intermediate, coarse for core layers). The three-layer particleboard is most commonly manufactured in the wood composite industry. This type of particleboard consists of a core layer and two face layers. The size of the core particles is coarser than that of the face particles. The coarse core materials provide the bulk of particleboard and the fine face materials provide smooth surfaces for laminating, painting, overlaying, or veneering. Most applications for particleboard require smooth surfaces (Vansteenkiste, 1981).

In addition, surface quality of particleboards can be dramatically improved by applying very thin overlays such as veneers, laminates, melamine impregnated papers, and vinyl films on the particleboard surfaces (Hiqiroglu and Suchsland, 1993). The surface overlays have to be smooth and can withstand the resistance stress of peeling. The rough surfaces of either the particleboard or the overlays decrease the contact between the particleboard and the overlays, which may result in surface irregularities, weak glue lines and low bonding strength (Nemi et al., 2005). Surface irregularities of particleboard would lower the product grade.

Many plant-based materials have been used for making particleboard. These materials include wheat straw (Cheng et al., 2004: Mo et al., 2003), waste tea leaves (Yalinkilic et al., 1998), kiwi and wine pruning (Nemli et al., 2003; Ntalos and Grigoriou, 2002), Norway spruce, Scots pine (Boonstra et al., 2006), needle litter of *Pinus pinaster* Ait. (Nemi and Ayadin, 2007), *Pinus radiate*, ponderosa and tanoak bark (Anderson et al., 1961; Hall et al., 1960), steam-treated rice industry residues (Geradi et al., 1998), saline Athel wood (Zheng et al., 2006), beech, pine, poplar (Nemli, 2007), cotton carpel (Alma et al., 2004), sunflower stalks (Gerjejansen, 1977; Khristova et al., 1998; Guler et al., 2006), rice husks (Vasisth and Chandramouli, 1975), durian peel and coconut coir (Kgedaru et al., 2004) and kenaf (Grigoriou et al., 2001). Particleboard can be made with lower grade plant materials than other wood composite panels such as OSB, plywood, and glulam. In addition, recycled woody materials are widely used for manufacturing particleboard. All of plant materials can essentially be utilized for making particleboard without any waste.

Another advantage of particleboard is its very broad application. About one-third of the particleboard goes into making kitchen and stereo/TV cabinets (Haygreen and Bowyer, 1996). Other common applications for particleboard include stair treads, home structures, table and counter tops, shelving, domestic, institutional and office furniture, vanities, speakers, sliding doors, lock blocks, interior signs, displays, table tennis tables, pool tables, and electronic game consoles (Nemli et al., 2005). All these broad products are mainly used indoor because particleboard panels typically do not have high enough water-resistant properties for outdoor applications.

Development history of particleboard panels

In 1902, Ernst Hubbard published a paper, “the Utilization of Wood-Waste.” This was the first publication about making particleboard. The particleboard panels were made by hot-pressing a mixture of sawdust and blood albumen. In 1905, Watson, an American inventor, invented a modern flakeboard by using wood particles. In 1914, Carl G. Muench produced particleboard panels using a technology similar to papermaking. In 1940, Humble found a way to utilize large volumes of sawdust and planer shavings in Germany. In 1941, during the World War II, one of the factories in Bremen, Germany started using spruce chips and phenolic adhesives for manufacturing particleboard panels. Farley & Loetscher Mfg Co., an American manufacturer, also started to produce the particleboard in the US during the same period of time. At that time, the particleboard market grew rapidly in Europe and the US.

By the end of the 1940s, logs for making plywood were on a short supply in Europe. Hence, some plywood was substituted by particleboard. In the mean time, in the North America, there were a large amount of sawmill wastes, which stimulated the production of particleboard. The early typical process for manufacturing particleboard used hammermill to break wood into small particles.

In the 1960s, the production of particleboard grew significantly with the innovation of hot press and resin technology. The Willamette Industries constructed in 1960 a new particleboard plant in Albany, Oregon to produce a new type of particleboard panels called “Duraflake” or “three-layer particleboard” that laid out fine particles in the outer layers and coarse particles in the inner layer. The Duraflake presented smooth outer surfaces and was durable, aesthetic, cost-competitive, and of large size, which enabled it to dominate the furniture market at that time. The most commonly used wood for the particleboard manufacture in the northern part of the U.S. was Douglas fir. The low pH level in Douglas fir facilitated the cure of UF resins used for making the particleboard (Maloney, 1977).

Particleboard was later classified according to its density: high density, medium density and low density. The property requirements, as specified by the American Nationals standard for particleboard A208.1 (ANSI/A208.1, 1999) for various grades of particleboard and particleboard flooring products are shown, respectively, in table 1 and 2.

Table 1: The property requirements, as specified by the American Nationals standard for Particleboard A208.1 (ANSI/A208.1, 1999)for various grades of particleboard products.

Grade[a]	MOR (MPa)	MOE (MPa)	Internal Bonding (Mpa)	Hardness (N)	Linear expansion max. avg (%)	Screw-holding (N)		Formaldehyde maximum emission (ppm)
						Face	Edge	
H-1	16.5	2400	0.90	2225	NS	1800	1325	0.30
H-2	20.5	2400	0.90	4450	NS	1900	1550	0.330
H-3	23.5	2750	1.00	6675	NS	2000	1550	0.30
M-1	11.0	1725	0.40	2225	0.35	NS	NS	0.30
M-S	12.5	1900	0.40	2225	0.35	900	800	0.30
M-2	14.5	2225	0.45	2225	0.35	1000	900	0.30
M-3	16.5	2750	0.55	2225	0.35	1100	1000	0.30
LD-1	3.0	550	0.10	NS	0.35	400	NS	0.30
LD-2	5.0	1025	0.15	NS	0.35	550	NS	0.30

[a] From NPA (1999). Particleboard made with phenol-formaldehyde-based resins does not emit significant quantities of formaldehyde. Therefore, such products and other particleboard products made with resin without formaldehyde are not subject to formaldehyde emission conformance testing.

[b] Panels designated as "exterior adhesive" must maintain 50% MOR after ASTM D1037 accelerated aging.

[c] MOR = modulus of rupture: MOE = modulus of elasticity. NS = not specified. 1 MPa = 145 lb/in^2; 1 N = 0.22 lb.

[d] H = density > 800 kg/m^3 (> 50 ;b/ft^3), M = density 640 to 800 kg/m^3 (> 40 to 50 lb/ft^3), LD = density < 640 kg/m^3 (< 40 ;b/ft^3), Grade M-S refers medium density; "special" grade added to standard after grades M-1, M-2, and M-3. Grade M-S fails between M-1 and M-2 in physical properties.

Table 2: The property requirements, as specified by the American Nationals standard for Particleboard A208.1 (ANSI/A208.1, 1999)for various grades of particleboard flooring products.

Grade[b]	MOR (MPa)	MOE (MPa)	Internal bond (MPa)	Hardness (N)	Linear expansion max avg (%)	Formaldehyde maximum emission (ppm)
PBU	11.0	1725	0.40	2225	0.35	0.20
D-2	16.5	2750	0.55	2225	0.30	0.20
D-3	19.5	3100	0.55	2225	0.30	0.20

[a]From NPA (1999). Particleboard made with phenol-formaldehyde-based resins does not emit significant quantities of formaldehyde. Therefore, such products and other particleboard products made with resin without formaldehyde are not subject to formaldehyde emission conformance testing. Grades listed here shall also comply with appropriate requirements listed in section 3. Panels designated as “exterior adhesive” must maintain 50 % MOR after ASTM D1037 accelerated aging (3.3.3)

[b]PBU = underlayment; D = manufactured home decking.

The particleboard manufacturing process

A typical particleboard manufacturing process begins with the preparation of wood particles. Wood or other plant materials are typically processed into particles with appropriate sizes using a hammermill in a particleboard plant. The ideal wood particles for making particleboard are long and thin and have similar sizes (Youngquist, 1999). The desirable size of particles normally depends on the types of particleboard. Core particles of three-layer particleboard are longer and thicker than surface particles. A graduated particleboard is different from the three-layer particleboard and can use diverse sizes of particles.

Wood particles are screened to remove very fine particles and oversized particles and to separate the core particles from the surface particles. The reason for removing very fine particles is that they have very large surface area and consume a high amount of an adhesive. Oversized particles are often the weak parts of particleboard and have to be removed for providing uniform properties. Either vibrating screen or gyratory screen (air stream-classifying screens) can be used for separating the core and the face materials. The most commonly used gyratory screen consists of wire cloth, and plates with holes or slots. Classified wood particles will be conveyed to storage bins.

Mechanical conveyer and air conveyer (pneumatic transport) are typically used for transporting small particles. The selection of these conveyers depends on the size of particles. The air conveyer forces particles to pass through the fans and can further reduce the sizes of particles, especially those that are oversized.

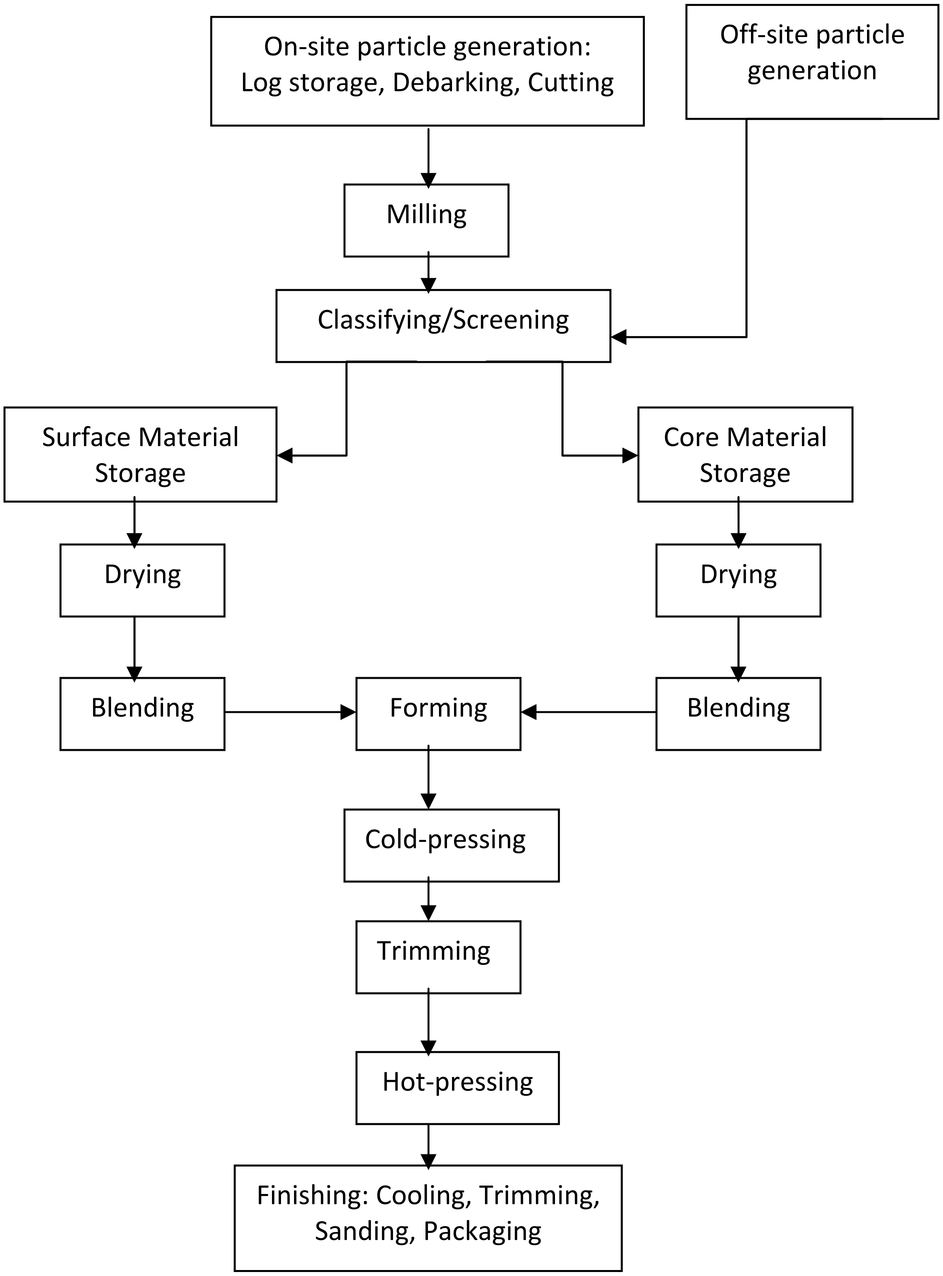
Wood particles in the storage bins are transferred to a dryer for reducing the moisture content. The moisture content of wood particles can range from 10% to 200%, and has to be reduced to about 2% to 7%. The most commonly used dryer in particleboard plants is a rotary dryer. The temperature in a dryer depends upon the moisture content of inlet particles. When wet particles are used, the dryer inlet temperature can be as high as 1600 °F. For relatively dry particles, the inlet temperature usually is not higher than 500 °F. Because the core particles have to be drier and have bigger sizes than the face particles, the dryer for drying the core particles will be operated at a higher temperature or for a longer period of time than that for drying the face particles. The moisture content of particles is vital for hot-pressing (Youngquist, 1999). Particleboard panels might be blown-up or have low bonding strengths if the moisture content is excessive. The internal bonding strength is also low if the particles are too dry. Therefore, the drying temperature and drying time have to be carefully selected for different hot-press and different particleboard products.

After drying, the particles are blended with an adhesive and waxes. The adhesive and waxes are sprayed onto the particles through a spraying nozzle, commonly called atomizer. The commonly used adhesive is urea–formaldehyde (UF) resins. Phenol–formaldehyde (PF) resins are also used for the production of particleboard panels. The PF-bonded particleboard panels are typically used for exterior applications. Formaldehyde-scavenger and a catalyst are typically included in the resins. In addition to the adhesive, addition of paraffin or microcrystalline wax is necessary for providing particleboard panels with short-term moisture-resistance. In addition, wax can increase the dimensional stability of the final product under wet conditions and decrease the possibility of equipment plugging. The usage of wax is approximately 0.3 to 1% of the oven-dry weight of the particles (Youngquist, 1999).

The adhesive-coated wood particles are formed into an even and consistent mat over a moving convey belt. The mat-formation can be batch-wise or continuous. The mat is typically cold-pressed to reduce mat thickness. In the continuous process, the particles are distributed onto a moving belt, and the amount of distributed particles is volumetrically controlled. After cold-press, the mat is cut into a designated size and transferred into a hot-press.

The hot-press is a very important step of making particleboard panels. The hot-pressing brings the particle surfaces together for bonding, and provides heat to cure the thermosetting adhesive. The hot-pressing also controls the thickness and thus the density of the final particleboard products. There are two types of hot-press machines: batch hot-press and continuous hot-press. The batch hot-press has multiple openings and makes multiple particleboard panels at the same time. The continuous hot-press has only one-opening, but the mat is moving. i.e., particleboard is made continuously. The total press time is around 150 seconds for a continuous hot-press and can be up to 300 seconds for a multi-opening batch press. The operating temperature is ranging from 300 °F to 360 °F. The moisture content of the mat before hot-press is typically about 8 to 12%. Hot-press is typically heated by steam generated from a wood-burning boiler in the particleboard plant.

After hot-pressing, particleboard panels go into a finishing step which includes trimming, sanding, packaging, and shipping. The edges of particleboard panels are trimmed to designed length and width. The trimming of a particleboard panel normally losses around 0.5 – 8% of its weight depending upon the panel size, and the process employed. The trimmed materials can be reused for making particleboard. A standard trimmer consists of saws with tungsten carbide tips. The particleboard sometimes needs to be sanded to get smooth surfaces. The schematic diagram of a particleboard manufacturing process is illustrated below.


On-site particle generation: Log storage, Debarking, Cutting
Off-site particle generation
Milling
Classifying/Screening
Surface Material Storage
Core Material Storage
Drying
Drying
Blending
Forming
Blending
Cold-pressing
Trimming
Hot-pressing
Finishing: Cooling, Trimming, Sanding, Packaging

Wood adhesives

Adhesive is a substance capable of holding materials together by surface attachment (Blowquist, 1999). An assembly formed with an adhesive consists of two objects, adherends, connected to each other by a layer of an adhesive material. Adhesion is the state held two surfaces together by some forces, which are interfacial forces, physical attraction, and chemical bonding (Vick, 1999).

Mechanical interlocking may have the main effect on bonding porous structure together, resulting from the penetration of an adhesive into materials and then solidifying of the adhesive. The most efficient mechanical interlocking is when the adhesive penetrates into the wood around two to six cells deep. An increase in the surface area for the molecular interaction between an adhesive and wood fibers is the most effective way of enhancing the mechanical interlocking forces (Gollob and Wellons, 1990).

Van der Waals forces play important roles in wood bonding. The Van der Waals forces include dipole-dipole interactions and hydrogen bonding. Hemicelluloses and cellulose are rich in hydroxyl groups for forming hydrogen bonding. Hence, the hydrogen bonding could make a major contribution to the adhesive strength of wood (Pizzi, 1994). Currently, many people believe hydrogen bonding is the major adhesion force in wood composites.

There may exist covalent bonds between the adhesive and wood substrates. However, there is no specific proof that covalent bonds play an important role in the strength of wood composites (Vick, 1999).

History of wood adhesives

Wood adhesives have played a key role in the growth and continued success of the forest product industry and have been a main solution in the efficient utilization of woody materials. Starch-based and protein-based adhesives were the early wood adhesives for bonding wood (Lambuth, 1983). The use of soy protein as an adhesive can be dated back to ancient time. However, its first commercial application as a wood adhesive did not start until 1920s (Brown, 2005). The soy protein was modified with lime and metallic salts for improving moisture-resistance and microbial resistance of the resulting wood composites (Blomquist, 1984). The substantial commercial use of soy-based adhesives has been documented from 1930s to 1960s. Nonetheless, the wood composites bonded with soy-based adhesives have a relatively lower strength and lower water resistance than those bonded with petroleum-based adhesives (Liu and Li, 2002).

Phenol–formaldehyde (PF) resins first appeared in a film form, which had limited applications for making wood composite panels. Its widespread use as a wood adhesive did not occur until its liquid form was developed in 1950s (Lambuth, 1983). Only a few years after the widespread application of PF resins in the wood composites market, urea-formaldehyde (UF) resins was also developed and used for making plywood. Resorcinol-formaldehyde (RF) resins were a newer wood adhesive than PF and UF resins. RF resins were extensively used for manufacturing military equipment during the World War II.

Synthetic petroleum-based resins quickly replaced natural material-based adhesives soon after they were successfully used in commercial production of wood composite panels because the resulting panels had superior mechanical properties and moisture-resistant properties. At present, synthetic resins dominate the wood adhesive market. In 1998, the North America wood composites industry consumed 1.78 million of solid resins. Only 0.3% of overall resins were derived from the soy-based products (Sellers, 2001).

Types of wood adhesives

The wood adhesives can be basically categorized into natural adhesives and synthetic adhesives.

Natural adhesives

Plant-material-based adhesives

Soybeans have been used as diets for around 5000 years. They have not been developed as adhesives until 1920s, (Sellers, 1985). Soy-based wood adhesives had many excellent properties such as low press temperatures, the ability to bind wood with relatively high moisture content, ease of handling, and low cost. The soy-based adhesives were more durable than starch-based adhesives, but were less moisture-resistant than casein-based adhesives. Therefore, soy-based adhesives were primarily used for making interior-graded plywood panels.

Another major plant-material-based adhesive is derived from starch. Starch is very abundant and inexpensive. Starch is a very strong adhesive for cellulosic materials such as paper because starch has abundant hydroxyl groups that can easily form hydrogen bonds and the Van der Waals interactions with wood components. Starch-based adhesives are presently widely used for making many paper products such as paper bags, paperboards, tapes, corrugated boxes, textiles, labels, envelopes, and wallpapers. However, the starch-based adhesives cannot be used for making wood composite panels because of their poor water-resistance.

Animal adhesives

The earliest adhesives that people used were animal glues. They derived from the collagen of animal skins, bones and tissues (Sellers, 1985). Proteins or polypeptides are the major components of animal glues. The adhesive properties such as viscosity and adhesive strengths are strongly dependent on the interactions of functional groups of amino acids (Subramanian, 1983). The properties of animal glues also depend upon the source where they are derived.

Animal glues can be obtained from boiling animal skins or animal bones in water. The resulting sticky solution was widely used for binding the small woody parts. Animal glues are still used in some furniture, crafting products, and paper tapes. Nevertheless, the quick gelation of the animal glues prevents them from being used in production of plywood. The availability of these protein-based adhesives is highly dependent upon the meat processing industry and hard to meet the huge demands of the wood composite industry (Sellers, 1985).

Other widely used animal glues are derived from casein, a milk protein. Casein has a much higher content of carboxylic acid groups than proteins from collagen. For this reason, casein is easily dissolved in an alkaline solution. Commonly used casein-based adhesives are called as casein-lime glues because they are prepared from reactions of casein and calcium hydroxide solution. Divalent calcium crosslinks the carboxylic acid groups in the casein to form water-insoluble calcium caseinate. Other polyvalent metal ions are able to work as cross-linkers. The adhesives can provide relatively strong joints, but are not very water-resistant. For instance, wood composites bonded with casein-lime glues might lose their integrity as the moisture content of the wood composites reaches 18% and remains at this level for some period of time (Subramanian, 1983). The casein-lime glues were mainly used for bonding joints in assemblies such as arches, doors, and beams before they were replaced by synthetic resins.

Protein-based adhesive can be derived from animal blood. The adhesive was prepared by spray-drying the soluble blood of domestic animals. This preparation process needed specialized equipments which were available at no more than a dozen meatpacking facilities in the U.S. (Sellers, 1985). Proteins from animal blood contain a high amount of polar functional groups, but their structures are globular. For achieving the full adhesive strength, the proteins have to be unfolded by dispersing them in an alkaline solution because amino acids in blood proteins are tightly held together through intermolecular interactions. The blood adhesives were ever used for the production of plywood. Combinations of blood protein and soy protein appeared to be better than blood adhesives or soy adhesives in terms of improving the water-resistance of the resulting wood composites (Subramanian, 1983). However, blood adhesives have been completely replaced by synthetic resins because of the low strength, low water-resistance of the resulting wood composite panels.

Synthetic resins

There are two types of synthetic resins: thermoplastic resins and thermosetting resins. The differences between thermoplastic resins and thermosetting resins are their chemical structures and responses to heat.

Thermoplastic resins

Thermoplastic resins are polymers that can soften or melt when heated, and solidify when cooled down. The softening and solidifying behaviors are reversible; hence, thermoplastic polymers can be melted and solidified many times without degradation (Eckelman, 1997). When used as adhesives, they are called hot-melt adhesives. Mechanical interlocking is the key adhesion mechanism. In the use of hot-melt adhesives, organic solvent is not involved and little volatile organic compounds are emitted. Environmental pollution is thus not a concern for hot-melt adhesives. The biggest drawback of hot-melt adhesives is their thermoplastic property. Wood composite panels bonded with hot-melt adhesives cannot be used in hot environment. Hot-melt adhesives used in wood composite panels can be derived from polyamides, poly(ethylene-vinyl acetate), polyvinylacetate, and polyacrylate.

The polyamide-based hot-melt adhesives are used in furniture manufacturing, and the poly(ethylene-vinyl acetate)-based adhesives are extensively used in book-binding, carpet, and shoe making. Poly(ethylene-vinyl acetate) is a random copolymer, its adhesive properties depend upon the ratio of ethylene and vinyl acetate.

Poly(vinyl acetate) (PVA) or “White glue” is another widely used wood adhesive. PVA is typically prepared from emulsion polymerization of vinyl acetate. PVA provides durable and invisible glue lines, and can provide strong bonding for cellulosic materials, especially wood. PVA is commonly used in furniture assembly, paper gluing, and wood working. Nonetheless, poor gap-filling, water-, and heat-resistant properties limit the application of PVA.

Polyacrylate-based adhesives, simply called acrylic adhesives are also used for bonding wood. Acrylic adhesives can be used by *in situ* polymerization of acrylate monomers. For example, acrylic adhesives are used to tightly fasten wooden nuts and bolts. Air has to be excluded for this application because oxygen in air inhibits the polymerization. The best known example of this acrylic adhesive is Super Glue. Super Glue consists of highly reactive α-cyanoacrylate that rapidly polymerize by moisture or water adsorbed on adherend surfaces (Subramanian, 1983).

Thermosetting resins

Thermosetting resins cross-link while heating, and cannot go back to their original chemical structures while cooling. Phenol-formaldehyde (PF) resins and urea-formaldehyde (UF) resins are the major thermosetting resins used in the production of wood composites.

PF resins are prepared from polymerization of phenol and formaldehyde. The adhesive properties of the PF resins highly depend upon the molar ratio of phenol to formaldehyde as well as a catalyst used in the reaction. Alkali-catalyzed PF resins are called resole, and acid-catalyzed PF resins are called novolak. Resole can be directly used for bonding wood, whereas an additional amount of formaldehyde has to be added to novolak before being used for bonding wood. The PF resins are extensively used for the production of exterior structural wood composites such as laminated veneer lumber (LVL) and softwood plywood because the resulting wood composites are strong, durable and water-resistant even under severe weather conditions. The PF resins in making wood composites are typically cured at 130°C-150°C for a few minutes. The PF resins have brownish to dark color after cured, which makes wood composite panels bonded with the PF resins not being aesthetic for interior application.

UF resins are most commonly used adhesives for making wood composition panels. The UF resins are prepared from polymerization of urea and formaldehyde. The molar ratio of formaldehyde to urea is typically around 1.5 to 2. Structures (Linear vs. branched) and molecular weights of the UF resins are highly dependent upon the formaldehyde/urea ratio and reaction conditions. The UF resins are cured under slightly acidic conditions. The commonly used catalysts for curing the UF resins include ammonium chloride, ammonium sulfate, and some organic acids such as tartaric acids and citric acids (Subramanian, 1983). The UF resins are water-soluble, light in color, inexpensive, non-flammable, and easy to cure. Approximately 85% of the UF resins is used in the production of wood composites. Interior grade plywood, medium density particleboard (MDF), and particleboard are almost exclusively made with the UF resins.

Other thermosetting adhesives such as polyurethanes, epoxy resins, and isocyanates are also used in the wood composite manufacturing.

Classification of adhesives

Wood adhesives are sometimes classified in accordance with water-resistant requirements and applications of the resulting wood composite panels. If the wood composite panels are for interior application, i.e., are used indoor, the adhesives used are called interior wood adhesives or interior grade wood adhesives. If the wood composite panels are for exterior application, i.e., are used outdoor, the adhesives used are called exterior wood adhesives or exterior grade wood adhesives. The interior wood adhesives are subdivided into interior moisture-resistant adhesives and interior water-resistant adhesives.

Wood composite panels bonded with interior moisture-resistant adhesives are intended to be used in low humidity environment, and are not intended for applications where they are exposed to high humidity or have direct contact with water. Some UF resins, soy-based adhesives, casein and blood glues are typically classified as interior moisture-resistant adhesives (Sellers, 1985).

Wood composite panels bonded with interior water-resistant adhesives are intended for applications where they may be exposed to high humidity or have direct contact with water temporarily. They are also required to retain their properties after temporary exposure to some other severe environments such as high temperature (over 50°C) and direct sunlight exposure. However, they are not required to withstand under these harsh conditions for a long period of time. Melamine is commonly incorporated in UF resins for improving their water-resistance. Most UF resins and all melamine-urea-formaldehyde (MUF) resins are interior water-resistant wood adhesives.

Wood composite panels bonded with exterior wood adhesives are intended for applications where they may have direct contact with water for a long period of time. These wood composite products are also expected to be resistant to microbial degradation, sunlight degradation and thermal degradation. PF resins, resorcinol-formaldehyde (RF) resins, and phenol-resorcinol-formaldehyde (PRF) resins are typically classified as exterior wood adhesives.

Issues associated with synthetic resins

Phenol is derived from petrochemical benzene and formaldehyde from methanol that is prepared from natural gas. In other words, raw materials for making UF and PF resins are mainly derived from non-renewable petroleum and natural gas. The reserve of fossil oil and natural gas is finite. Petroleum and natural gas are mainly used of fuel energy. The fuel consumption increases as the world's population grows, which will increase the prices and availability of raw materials of wood adhesives (Sellers, 1985). The growth and continued success of the wood composite industry will benefit if wood adhesives are derived from renewable materials such as soy proteins.

Emission of formaledyde from wood composite panels bonded with UF resins was first recorded in 1962 (Marutzky, 1989). Since then, the emission of formaldehyde from wood composite panels bonded with various UF resins has been extensively investigated. It has been known for a long time that formaldehyde is hazardous to human health such as causing skin, eye and throat irritations as well as respiratory discomfort. After long-term study, International Agency for Research on Cancer, a well respected research institute of World Health Organization, re-classified formaldehyde from probable carcinogenic to carcinogenic to human in 2004. California Air Resources Board (CARB) estimated that wood-based products released about 400 tons of formaldehyde in California every year and issued a strict regulation in 2007 on lowering formaldehyde emission from wood-based products used and sold in California (Lent, 2006).

Several Formaldehyde-free adhesives have been developed from renewable materials such as soy flour (Li, 2004; Li, 2005; Liu, 2007). One of the formaldehyde-free soy-based adhesives has been used for commercial production of decorative hardwood plywood since 2004.

Reference

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A New Method of Making Particleboard with a Formaldehyde-free Soy-based Adhesive

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ABSTRACT

A soy-based formaldehyde-free adhesive consisting of soy flour (SF) and a curing agent (CA) has been successfully used for the commercial production of interior plywood. However, this adhesive cannot be easily sprayed onto wood particles for making particleboard because of its high viscosity. In this study, a new method of using this soy-based adhesive for making particleboard was developed and investigated in detail. The procedure of making particleboard with this new method included: the coating of wood particles with soy slurry in water, the oven-drying of the wet soy-coated wood particles, the spraying of the CA onto the oven-dried soy-coated wood particles, and formation of a particle mat with the CA-soy-coated wood particles, and the hot-pressing of the mat into particleboard. Effects of particleboard density, adhesive usages for both core and face particles, the solids content of the soy slurry, hot-press time, hot-press temperature, the storage time of the wet soy-coated wood particles, and the CA/SF weight ratio on the internal bond strength (IB), the modulus of rupture (MOR), and the modulus of elasticity (MOE) of the resulting particleboard were investigated in detail. The optimal conditions for making particleboard in terms of enhancing the IB, MOR and MOE were: 760 kg/m^3 of the particleboard density, 11 wt% resin usage for the core particles, 12 wt% resin usage for the face particles, 20 wt% solids content of the soy slurry, 180 °C of the hot-press temperature, 224 s of the hot-press time, 1:7 CA/SF weight ratio, and 36 h of the storage time for the wet soy-coated wood particles. The strengths of particleboard made with this new method exceeded the industry property requirements for the M-2 particleboard.

KEYWORD

Wood adhesive, soy protein, particleboard, mechanical properties

1. Introduction

Particleboard panels are made with wood particles and a wood adhesive. Particleboard panels are extensively used for the production of furniture, floor underlayment, cabinets, home construction, tabletops, interior signs, kitchen countertops, electronic game consoles, table-tennis tables, bookshelves, office desk and many other products (Anon, 1996). The particleboard panels with a density of 640 kg/m^3 to 800 kg/m^3 are designated as medium-density particleboard (ASTM D1554-86, 1995). The medium density particleboard is the most commonly used particleboard. Urea-formaldehyde (UF) resins are predominately used for the production of particleboard (Zucaro and Reen, 1995). However, there are two issues associated with UF resins. First, UF resins are derived from non-renewable petrochemicals. The sustainable growth and continued success of particleboard industry are dependent upon the development of new adhesives from renewable materials. The second issue is that carcinogenic formaldehyde is released in the production and use of particleboard bonded with UF resins (Mo et al., 2003), (Sauter, 1996) (Brown, 2005). The California Air Resource Board has issued a tough regulation of limiting formaldehyde emission from wood-based products sold and used in California in 2007. The most desirable way of resolving these two issues is to use formaldehyde-free adhesives from renewable resources (Huang, 2008)

Several formaldehyde-free wood adhesives from renewable materials such as soy flour have been developed. One of the formaldehyde-free soy-based adhesives has been used for the commercial production of plywood since 2004 (Li, 2004). The soy-based adhesive used in the commercial production of decorative plywood includes two basic ingredients: defatted SF and a small amount of a curing agent. This soy-based adhesive has a high viscosity. It is very difficult to spray the adhesive onto wood particles for making particleboard panels. For making particleboard with this adhesive, the curing agent (a dilute aqueous solution of a polymer) was sprayed onto a mixture of wood particles and defatted SF. Particleboard panels made with this method had good strengths. However, this method had a drawback that a significantly longer press time than the conventional industry process with the UF resins was required. A long press time means a low production rate and consequently a high price of particleboard panels.

For lowering the viscosity of the adhesive, soy flour was treated with chemicals such as urea before mixing with the curing agent (Wescott, 2008). This method allowed direct spraying of the adhesive onto wood furnishes. No data have been published on whether this method would produce good particleboard panels. The chemicals used for treating SF and the chemical modification process both add costs to the soy-based adhesive. The chemical modification also increases the transportation cost of the treated soy because the total solids content of the treated soy is only about 50%. Whoever does the chemical treatment would have to make some profit, which would further increase the cost of the soy-based adhesive.

In this project, we developed a new method of using the original recipe of the soy-based adhesive used in plywood production for making particleboard. SF was first mixed with water to form dilute soy slurry that can be easily mixed onto wood particles. The soy-coated wood particles were dried to certain moisture content and then further coated with an aqueous curing agent. The development of this technique and properties of the resulting particleboard panels are described in detail.

2. Experiments

2.1. Materials

Core wood particles and face wood particles of particleboard were provided by Flakeboard (Albany, OR). Soy flour (SF) with 7% moisture content, 100 mesh, and 90 PDI (protein dispersibility index) was from Cargill Incorporated (Minneapolis, MN), and CA1000 curing agent (CA) with 20 wt% solid content was received from Hercules Incorporated (Wilmington, DE).

2.2. Preparation of particleboard panels bonded with the soy-based adhesive

Soy flour (SF) (259 g) and water (935 mL) were mixed in a HOBART A-200 blender (Hobart, Topeka, KS) at room temperature at the maximum speed for 10 min to form a 20% soy slurry. Soy slurry with a different solids content was prepared through changing the SF/water weight ratio. Core wood particles (2665 g) were added to the resulting soy slurry in the same blender, and the resulting mixture was stirred for 5 min to obtain wet soy-coated particles where soy/wood particle weight ratio was 11%. Wet soy-coated particles with a different soy/wood particle weight ratio were prepared in the same way except the use of different amount of SF, water and wood particles. Wet soy-coated surface particles were prepared in the same way as the wet soy-coated core particles. The soy-coated particles were dried in a rotary dryer (Speedqueen, Ripon, WI) at 103°C for 46 min when the moisture content of the soy-coated particles was about 3-4%. The drying time was used to adjust the moisture content of the soy-coated particles.

Energy consumption of the rotary dryer obtained from $Q = P \cdot t$

Q represented the total energy consumption (MJ); P represented the energy consumption of rotary dryer per hour; t = time (h). The total energy consumption of dryer to lower the moisture content to 3-4% was 97 MJ

The CA was sprayed onto the dried soy-coated particles in a rotary blender. The CA-soy-coated particles were hand-formed into 508 mm x 508 mm mat. For the three-layer particleboard, the mat consisted of 35% face particles and 65% core particles (all on dry weight basis). More specifically, the CA-soy-coated face particles (863 g, 17.5% of the particleboard weight) were formed into a 508 mm x 508 mm mat with uniform thickness. The CA-soy-coated core materials (3121 g) were then uniformly laid on the top of this face layer, followed by a uniform layer of the CA-soy-coated face particles. The resulting mat was pressed at pre-determined pressure, temperature and time (see specific pressure, temperature and time at the figure legend of each figure) to form a particleboard panel. The resulting particleboard panel was left at ambient environment for 24 h before cutting. After trimming the edges of the panel, the final dimension of the particleboard panel was 444.5 mm x 469.9 mm and the moisture content was in a range of 6% to 7%. Two particleboard panels were produced for each experimental variable.

2.3. Evaluation of mechanical properties of the particleboard panels

Each edge-trimmed particleboard panel was cut into nine 50.8 mm x 50.8 mm test specimens for determination of internal bond strength (IB) testing by tensile test and four 76.2 mm x 469.9 mm specimens for determination of Modulus of Rupture (MOR) and Modulus of Elasticity (MOE). The MOR and MOE were obtained from a three-point bending test in accordance with ASTM D1037-99 (American Society for Testing and Materials, 1999). A MTS Sintech 1/G (MTS Systems Corp., Enum- claw, WA) testing machine was used for measuring the IB, MOR and MOE. The IB, MOR and MOE values were evaluated against the minimum industry requirements of M-2-grade particleboard (one of the most commonly used particleboard panels): IB= 0.45 MPa, MOR = 14.5MPa, and MOE = 2.225 GPa (ANSI/A208.1, 1999).

Table 1: The property requirements, as specified by the American Nationals standard for Particleboard A208.1 (ANSI/A208.1, 1999), for various grades of particleboard products.

Grade[a]	MOR (MPa)	MOE (MPa)	Internal Bonding (Mpa)	Hardness (N)	Linear expansion max. avg (%)	Screw-holding (N)		Formaldehyde maximum emission (ppm)
						Face	Edge	
H-1	16.5	2400	0.90	2225	NS	1800	1325	0.30
H-2	20.5	2400	0.90	4450	NS	1900	1550	0.330
H-3	23.5	2750	1.00	6675	NS	2000	1550	0.30
M-1	11.0	1725	0.40	2225	0.35	NS	NS	0.30

M-S	12.5	1900	0.40	2225	0.35	900	800	0.30
M-2	14.5	2225	0.45	2225	0.35	1000	900	0.30
M-3	16.5	2750	0.55	2225	0.35	1100	1000	0.30
LD-1	3.0	550	0.10	NS	0.35	400	NS	0.30
LD-2	5.0	1025	0.15	NS	0.35	550	NS	0.30

[a] From NPA (1999). Particleboard made with phenol-formaldehyde-based resins does not emit significant quantities of formaldehyde. Therefore, such products and other particleboard products made with resin without formaldehyde are not subject to formaldehyde emission conformance testing.

[b] Panels designated as "exterior adhesive" must maintain 50% MOR after ASTM D1037 accelerated aging.

[c] MOR = modulus of rupture: MOE = modulus of elasticity. NS = not specified. 1 MPa = 145 lb/in^2; 1 N = 0.22 lb.

[d] H = density > 800 kg/m^3 (> 50 lb/ft^3), M = density 640 to 800 kg/m^3 (> 40 to 50 lb/ft^3), LD = density < 640 kg/m^3 (< 40 lb/ft^3), Grade M-S refers medium density; "special" grade added to standard after grades M-1, M-2, and M-3. Grade M-S fails between M-1 and M-2 in physical properties.

2.4. Statistical analysis of experimental data

All experimental data with samples were analyzed with a standard two-sample t-test using S-PLUS statistical software (Version 8.0, Insightful Corp., Seattle, WA, USA). All comparisons were based on a 95% confidence interval.

3. Results

Effect of the particleboard density on the IB is shown in Fig. 1. The IB remained the same when the density was raised from 520 kg/m^3 to 560 kg/m^3, but significantly increased when the density was raised from 560 kg/m^3 to 680 kg/m^3. The IB flattened out when the density was further raised from 680 kg/m^3 to 760 kg/m^3.

The IB at all densities tested was the same or higher than the minimum industry requirement (0.45 MPa)

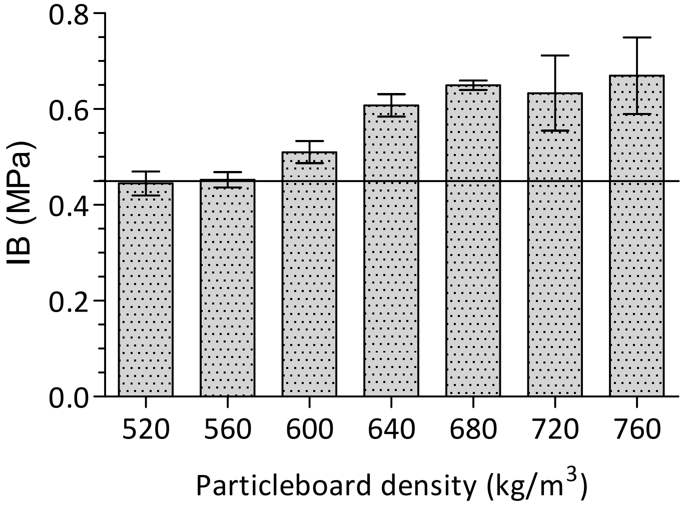

Fig. 1 Effect of particleboard density on the IB (the solids content of soy slurry, 20 wt%; the resin usage of the core particles, 11 wt%; the resin usage of the face particles, 12 wt%; the CA/SF weight ratio, 1/7; storage time of wet soy-coated wood particles, 30 min; hot-press temperature, 180 ºC, and hot-press time, 224 s). Each data point is the mean of 18 samples, and the error bar represents one standard deviation.

Effects of particleboard density on the MOR and MOE are shown in Fig. 2. The MOR slowly increased when the density was raised from 520 kg/m^3 to 640 kg/m^3, and then rapidly increased when the density was raised from 640 kg/m^3 to 720 kg/m^3. The MOR at 720 kg/m^3 was statistically the same as that at 760 kg/m^3. The MOR exceeded the minimum industry requirement (14.5 MPa, the horizontal solid line) when the density was at 680 kg/m^3 or higher. The MOE remained unchanged when the density was raised from 520 kg/m^3 to 560 kg/m^3, and the slightly increased from 560 kg/m^3 to 600 kg/m^3. The MOE fattened out from 600 kg/m^3 to 640 kg/m^3 and then gradually increased from 640 kg/m^3 to 720 kg/m^3. The MOE at 720 kg/m^3 was the same as that at 760 kg/m^3. All MOE values at the density range tested exceeded the minimum industry requirement (2.225 GPa, the horizontal dashed line) (Fig. 2). Results from Fig. 1 and 2 showed a general trend that the IB, MOR and MOE increased along with increasing the density. Therefore the density of 760 kg/m^3 was used in the subsequent investigation.

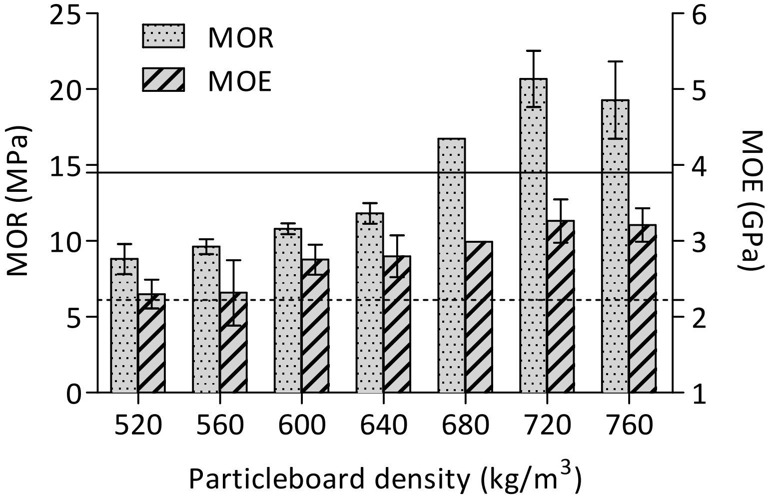

Fig. 2 Effect of particleboard density on the MOR and MOE (the solids content of soy slurry, 20 wt%; the resin usage of the core particles, 11 wt%; the resin usage of the face particles, 12 wt%; the CA/SF weight ratio, 1/7; storage time of wet soy-coated wood particles, 30 min; hot-press temperature, 180 ºC, and hot-press time, 224 s). Each data point is the mean of 8 samples, and the error bar represents one standard deviation.

Effect of adhesive usage of the core particles on the IB is shown in Fig. 3. The IB significantly increased when the adhesive usage was raised from 7% to 9%, and then slowly increased when the adhesive usage was further raised from 8% to 10%. The IB markedly increased when the adhesive usage was raised from 10% to 11%.

The IB at 10% or higher adhesive usage exceeded the minimum industry requirement of the M-2 particleboard (the horizontal solid line).

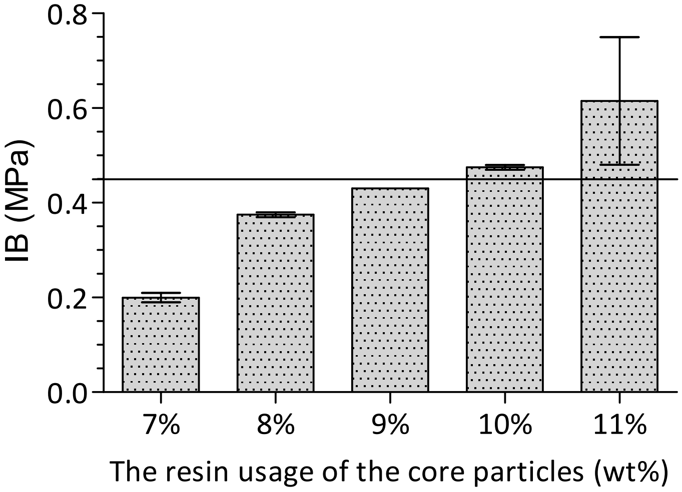

Fig. 3 Effect of the resin usage of the core particles on the IB (the solids content of soy slurry, 20 wt%; the resin usage of the face particles, 12 wt%; particleboard density, 760 kg/m^3; the CA/SF weight ratio, 1/7; the storage time of wet soy-coated particles, 30 min; hot-press temperature, 180 ºC; hot-press time, 224 s). Each data point is the mean of 18 samples, and the error bar represents one standard deviation.

Effect of adhesive usage of the core particles on the MOR and MOE is shown in Fig. 4. The MOR was statistically the same in the adhesive usage range of 7% to 9%. The MOR at 10% adhesive usage was somehow lowered than those at 9% and 11%. The MOR at 11% adhesive usage appeared to be the highest among all adhesive usages tested. The MOR values in the adhesive usage range tested met or exceeded the minimum industry requirement for the M-2 particleboard (horizontal solid line). The MOE at each adhesive usage level was statistically the same and exceeded the minimum industry requirement (horizontal dashed line) (Fig. 4).

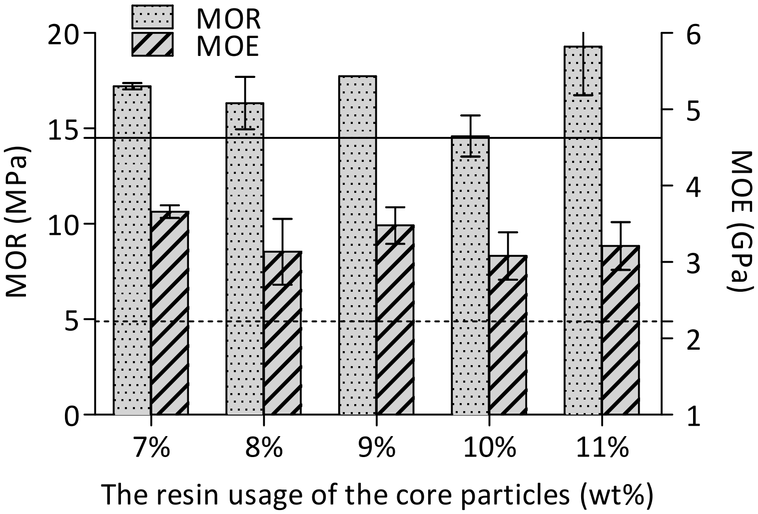

Fig. 4 Effect of the resin usage of the core particles on the MOR and MOE (the solids content of soy slurry, 20 wt%; the resin usage of the face particles, 12 wt%; particleboard density, 760 kg/m^3; the CA/SF weight ratio, 1/7; the storage time of wet soy-coated particles, 30 min; hot-press temperature, 180 ºC; hot-press time, 224 s). Each data point is the mean of 8 samples, and the error bar represents one standard deviation.

Effect of the adhesive usage of face particles on the IB is shown in Fig. 5. The IB was the same at the adhesive usage of 9% to 10% and then increased when the adhesive usage was raised from 10% to 12%. The further increase in the adhesive usage from 12% to 13% did not change the IB. The IB exceeded the minimum industry requirement (the horizontal solid line in Fig. 5) when the adhesive usage of face particles was 11% or higher.

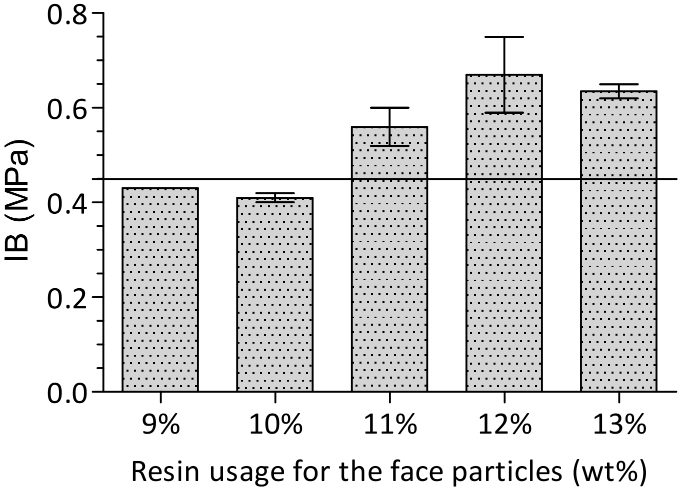

Fig. 5 Effect of the resin usage of the face particles on the IB (the solids content of soy slurry, 20 wt%; the resin usage of the core particles, 12 wt%; particleboard density, 760 kg/m^3; the CA/SF weight ratio, 1/7; the storage time of wet soy-coated particles, 30 min; hot-press temperature, 180 ºC; hot-press time, 224 s). Each data point is the mean of 18 samples, and the error bar represents one standard deviation.

Effects of the adhesive usage of face particles on the MOR and MOE are shown in Fig. 6. The MOR did not change when the adhesive usage was raised from 9% to 11% and then markedly increased when the adhesive usage was further raised from 11% to 12%. The MOR slightly decreased when the adhesive usage was raised from 12% to 13%. The MOR exceeded the minimum industry requirement (the horizontal solid line in Fig. 6) when the adhesive usage was 12% or higher. The average value of the MOE at each adhesive usage level fluctuated a bit and remained statistically the same in the whole adhesive usage range tested.

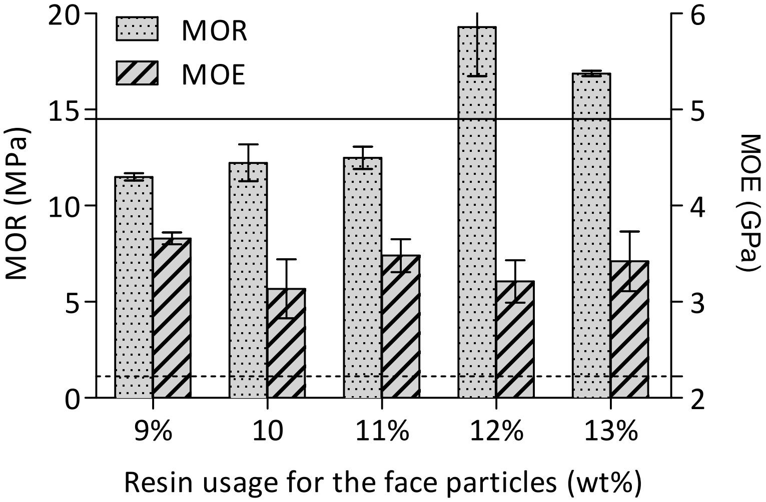

Fig. 6 Effect of the resin usage of the face particles on the MOR and MOE (the solids content of soy slurry, 20 wt%; the resin usage of the core particles, 12 wt%; particleboard density, 760 kg/m^3; the CA/SF weight ratio, 1/7; the storage time of wet soy-coated particles, 30 min; hot-press temperature, 180 ºC; hot-press time, 224 s). Each data point is the mean of 8 samples, and the error bar represents one standard deviation.

Effect of the solids content of soy slurry on the IB is shown in Fig. 7. The IB gradually increased when the solids content was raised from 5% to 20%. However the IB dramatically decreased when the solids content was further increased from 20% to 25%. The IB at the solids content of 5% to 20% met the minimum industry requirement of M-2 particleboard (the horizontal solid line).

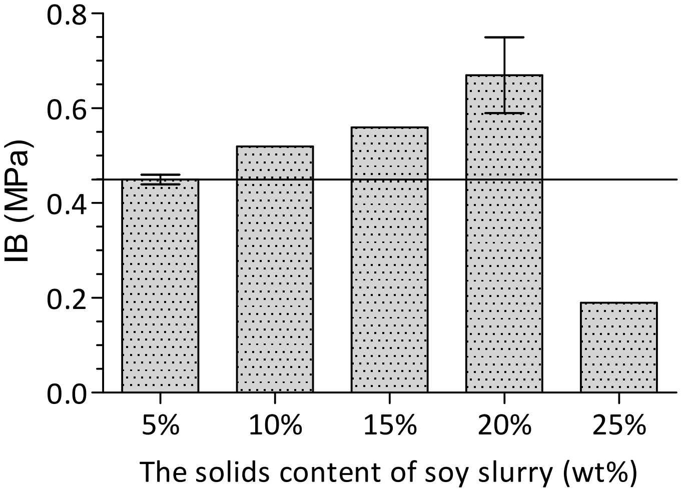

Fig. 7 Effect of the solids content of the soy slurry on the IB (the resin usage of the core particles, 11 wt%; the resin usage of the face particles, 12 wt%; particleboard density, 760 kg/m^3; the CA/SF weight ratio, 1/7; storage time of wet soy-coated particles, 30 min; hot-press temperature, 180 ºC; hot-press time, 224 s). Each data point is the mean of 18 samples, and the error bar represents one standard deviation.

Effects of the solids content of soy slurry on the MOR and MOE are shown in Fig. 8. The MOR gradually increased when the solids content was raised from 5% to 15%. The MOR at 15% was the same as that at 20%. The MOR markedly decreased when the solids content was further raised from 20% to 25%. When the solids content was in the range of 10% to 20%, the MOR exceeded the minimum industry requirement (the horizontal solid line). The MOE gradually increased when the solids content was raised from 5% to 15% and then gradually decreased when the solids content was further raised from 15% to 25%. The MOE in the solids content of 5% to

25% exceeded the minimum industry requirement (the horizontal dashed line) (Fig. 8).

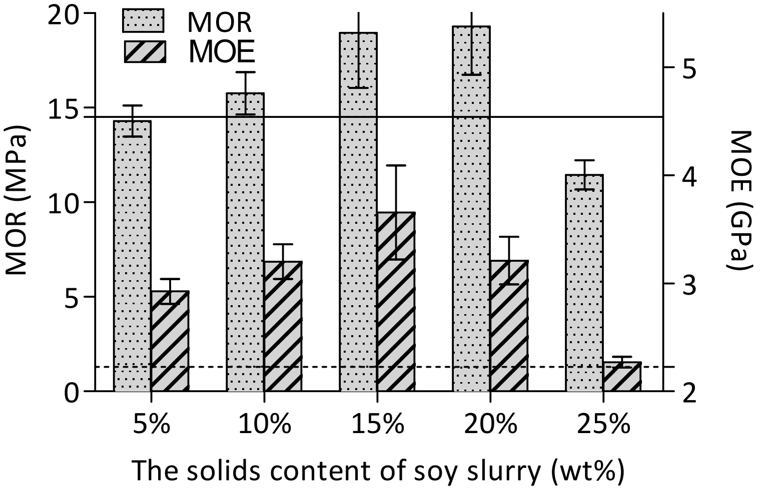

Fig. 8 Effect of the solids content of the soy slurry on the MOR and MOE (the resin usage of the core particles, 11 wt%; the resin usage of the face particles, 12 wt%; particleboard density, 760 kg/m^3; the CA/SF weight ratio, 1/7; storage time of wet soy-coated particles, 30 min; hot-press temperature, 180 ºC; hot-press time, 224 s). Each data point is the mean of 8 samples, and the error bar represents one standard deviation.

Effect of the hot-press time on the IB is shown in Fig. 9. The IB gradually increased when the hot-press time was raised from 183 s to 224 s. The IB at 245 s was slightly lower than that at 224 s. The IB markedly decreased when the hot-press time was further raised from 245 s to 266 s. The IB in the range of 203 s to 245 s exceeded the minimum industry requirement (the horizontal dashed line).

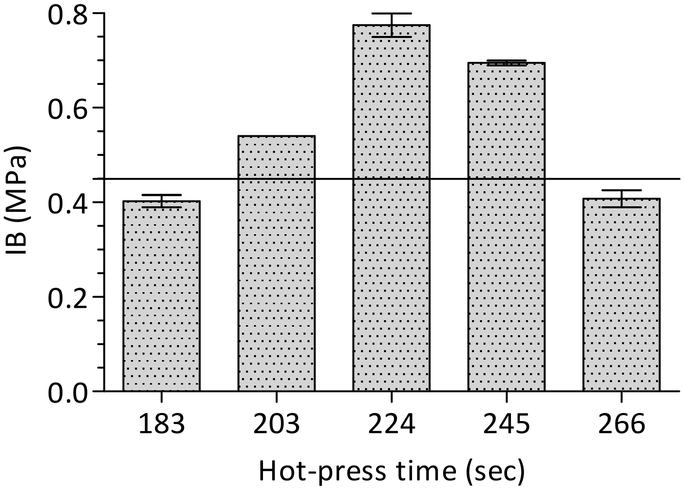

Fig. 9 Effect of the hot-press time on the IB (the solids content of soy slurry, 20 wt%; the resin usage of the core particles, 11 wt%; the resin usage of the face particles, 12 wt%; particleboard density, 760 kg/m^3; the CA/SF weight ratio, 1/7; storage time of wet soy-coated particles, 30 min; hot-press temperature, 180 ºC). Each data point is the mean of 18 samples, and the error bar represents one standard deviation.

Effects of the hot-press time on the MOR and MOE are shown in Fig. 10. The MOR at 203 s was somehow lower than those at 183 s and 224 s. The MOR at 224 s was the highest at all hot-press times studied. The MOR significantly decreased when the hot-press time was raised from 224 s to 245 s. However, further increasing the hot-press time from 245 s to 266 s did not significantly change the MOR. The MOR at all hot-press times except 203 s exceeded the minimum industry requirement (the horizontal solid line in Fig. 10). The MOE at all hot-press times studied (i.e., from 183 s to 266 s) was statistically the same and exceeded the minimum industry requirement (the horizontal dashed line in Fig. 10).

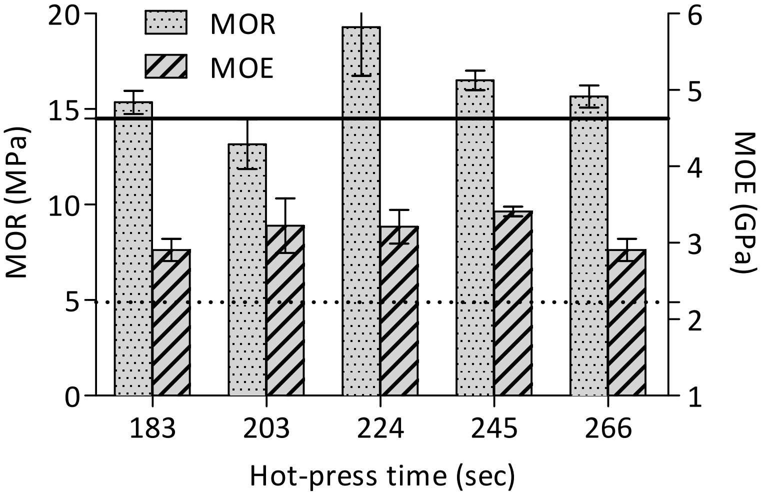

Fig. 10 Effect of the hot-press time on the MOR and MOE (the solids content of soy slurry, 20 wt%; the resin usage of the core particles, 11 wt%; the resin usage of the face particles, 12 wt%; particleboard density, 760 kg/m^3; the CA/SF weight ratio, 1/7; storage time of wet soy-coated particles, 30 min; hot-press temperature, 180 ºC). Each data point is the mean of 8 samples, and the error bar represents one standard deviation.

Effect of the hot-press temperature on the IB is shown in Fig. 11. The IB rapidly increased when the hot-press temperature was raised from 150 ºC to 180 ºC. However further increasing the hot-press temperature from 180 ºC to 190 ºC did not significantly change the IB. The IB at 180 ºC and 190 ºC exceeded the minimum industry requirement (the horizontal solid line in Fig. 11).

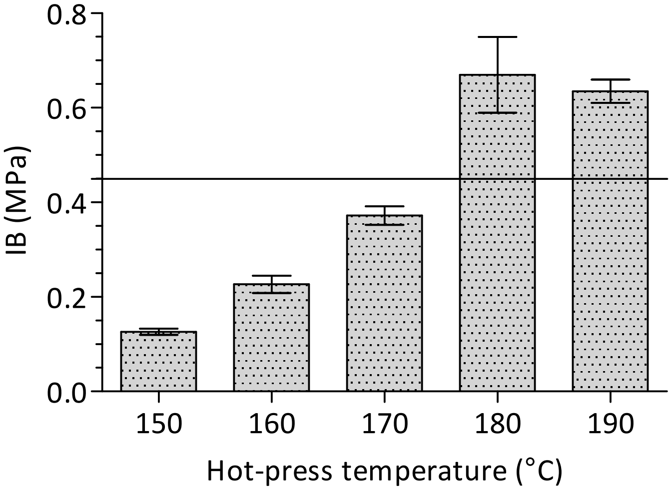

Fig. 11 Effect of the hot-press temperature on the IB (the solids content of soy slurry, 20 wt%; the resin usage of the core particles, 11 wt%; the resin usage of the face particles, 12 wt%; particleboard density, 760 kg/m^3; the CA/SF weight ratio, 1/7; storage time of wet soy-coated particles, 30 min; hot-press time, 224 s). Each data point is the mean of 18 samples, and the error bar represents one standard deviation.

Effects of the hot-press temperature on the MOR and MOE are shown in Fig. 12. The MOR rapidly increased when the hot-press temperature was raised from 150 ºC to 180 ºC and then significantly decreased when the hot-press temperature was further raised from 180 ºC to 190 ºC. The MOR at the hot-press temperatures of 170 ºC to 190 ºC exceeded the minimum industry requirement (the horizontal solid line in Fig. 12). The MOE rapidly increased when the hot-press temperature was raised from 150 ºC to 170 ºC. But the MOE at 180 ºC was significantly lower than those at 170 ºC and 190 ºC. The MOE at 170 ºC, 180 ºC and 190 ºC exceeded the minimum industry requirement (the horizontal dashed line in Fig. 12).

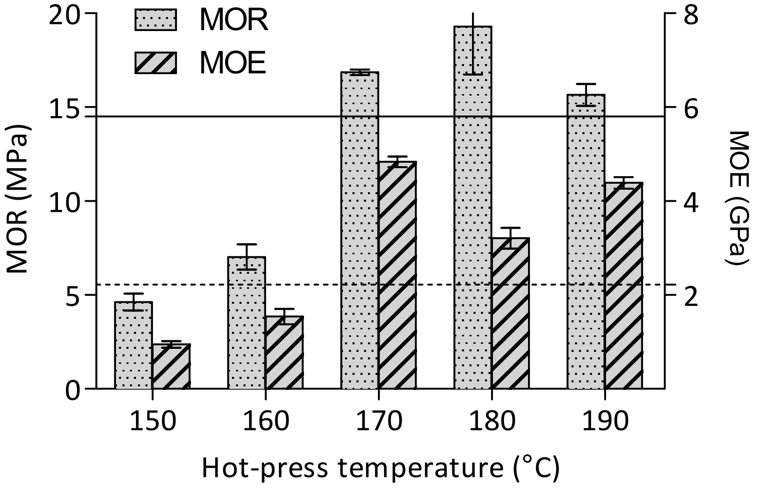

Fig.12 Effect of the hot-press temperature on the MOR and MOE (the solids content of soy slurry, 20 wt%; the resin usage of the core particles, 11 wt%; the resin usage of the face particles, 12 wt%; particleboard density, 760 kg/m^3; the CA/SF weight ratio, 1/7; storage time of wet soy-coated particles, 30 min; hot-press time, 224 s). Each data point is the mean of 8 samples, and the error bar represents one standard deviation.

Effect of the CA/SF weight ratio on the IB is shown in Fig. 13. The IB slightly increased when the CA/SF ratio was raised from 1/15 to 1/10, and markedly increased when the CA/SF ratio was raised from 1/10 to 1/7. Further increase in the CA/SF ratio from 1/7 to 1/5 did not significantly change the IB. However, the IB significantly decreased when the CA/SF ratio was increased from 1/5 to 1/4. The IB in the CA/SF ratio range of 1/10 to 1/4 exceeded the minimum industry requirement (the horizontal solid line in Fig. 13).

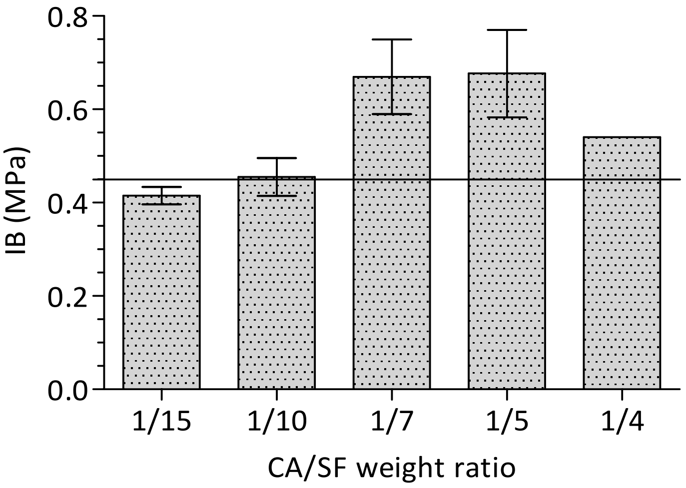

Fig. 13 Effect of the CA/SF weight ratio on the IB (the solids content of soy slurry, 20 wt%; the resin usage of the core particles, 11 wt%; the resin usage of the face particles, 12 wt%; particleboard density, 760 kg/m^3; storage time of wet soy-coated particles, 30 min; hot-press temperature, 180 ºC; hot-press time, 224 s). Each data point is the mean of 18 samples, and the error bar represents one standard deviation.

Effects of the CA/SF weight ratio on the MOR and MOE are shown in Fig. 14. The MOR gradually increased when the CA/SF ratio was raised from 1/15 to 1/7 and then gradually decreased when the CA/SF ratio was further raised from 1/7 to 1/4. The MOR in the CA/SF ratio range of 1/10 to 1/4 exceeded the minimum industry requirement (the horizontal solid line in Fig. 14). The MOR at the 1/15 CA/SF ratio was very close to meet the minimum industry requirement. The MOE rapidly increased when the CA/SF ratio was raised from 1/15 to 1/10, but remained the same when the CA/SF ratio was further raised from 1/10 to 1/5. The MOE markedly decreased when the CA/SF ratio was increased from 1/5 to 1/4. At all CA/SF ratios tested, the MOE exceeded the minimum industry requirement (the horizontal dashed line in Fig. 14).

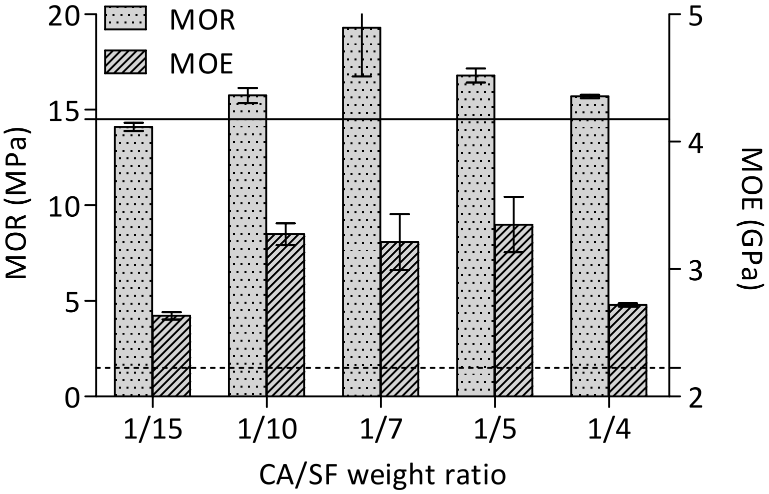

Fig. 14 Effect of the CA/SF weight ratio on the MOR and MOE (the solids content of soy slurry, 20 wt%; the resin usage of the core particles, 11 wt%; the resin usage of the face particles, 12 wt%; particleboard density, 760kg/m^3; storage time of wet soy-coated particles, 30 min; hot-press temperature, 180 ºC; hot-press time, 224 s). Each data point is the mean of 8 samples, and the error bar represents one standard deviation.

Effect of the storage time of the wet soy-coated wood particles on the IB is shown in Fig. 15. The IB did not significantly change when the storage time was in the range of 0.5 h to 36 h (Fig. 15). However, the IB markedly decreased when the storage time was raised from 36 h to 48 h (Fig. 15). The IB at all storage times tested exceeded the minimum industry requirement (the horizontal solid line in Fig. 15).

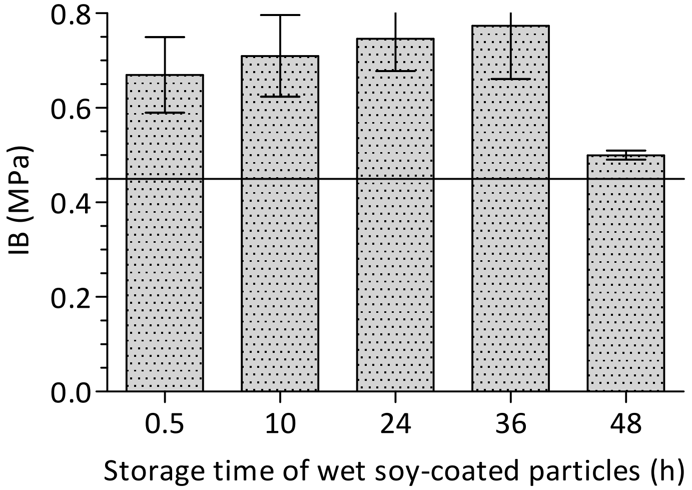

Fig. 15 Effect of the storage time of wet soy-coated particles on the IB (the solids content of soy slurry, 20 wt%; the resin usage of the core particles, 11 wt%; the resin usage of the face particles, 12 wt%; particleboard density, 760 kg/m^3; the CA/SF weight ratio, 1/7; CA/SF weight ratio, 1/7; hot-press temperature, 180 ºC; hot-press time, 224 s). Each data point is the mean of 18 samples, and the error bar represents one standard deviation.

Effects of the storage time of the wet soy-coated wood particles on the MOR and MOE are shown in Fig. 16. The MOR increased along with increasing the storage time from 0.5 h to 24 h. The MOR at 24 h storage time was the same as that at 36 h. The MOR at 48 h storage time was markedly lower than that at 36 h. At all storage times tested, the MOR exceeded the minimum industry requirement (the horizontal solid line in Fig. 16). The MOE gradually increased along with increasing the storage time from 0.5 h to 24 h, flattened out from 24 h to 36 h, and then decreased from 36 h to 48 h. The MOE at all storage times studied exceeded the minimum industry requirement (the horizontal dashed line in Fig. 16).

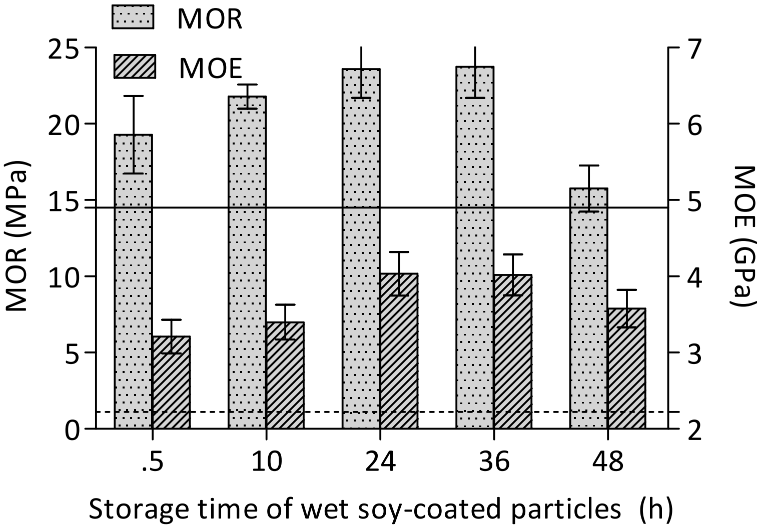

Fig. 16 Effect of the storage time of wet soy-coated particles on the MOR and MOE (the solids content of soy slurry, 20 wt%; the resin usage of the core particles, 11 wt%; the resin usage of the face particles, 12 wt%; particleboard density, 760 kg/m^3; the CA/SF weight ratio, 1/7; CA/SF weight ratio, 1/7; hot-press temperature, 180 ºC; hot-press time, 224 s). Each data point is the mean of 8 samples, and the error bar represents one standard deviation.

DISCUSSION

It is well established that the board density is one of the most important variables in determining the strengths of particleboard. In particleboard, the intimate contact among adhesive-coated wood particles is prerequisite for forming bonding among the particles. Raising the board density increases the intimate contact of the particles, thus increasing the strengths of the particleboard. However, if the density is high enough to allow all wood particles to intimately contact with each other, further increase in the density would not increase the strengths any more. The results shown in Fig. 1 and 2 are consistent with these explanations. For this particular adhesive, the board density has to be ≥ 680 kg/m^3 before the IB, MOR and MOE can all meet the minimum industry requirements for M-2 particleboard panels.

The wood surfaces must have a sufficient amount of the adhesive for providing good bonding, which explains the IB increase along the increase in the resin usage for the core wood particles (Fig. 3). The 11% resin usage appeared to be high enough for providing good internal bonding. A particleboard specimen typically failed in the core layer during the measurement of the IB. Therefore, the resin usage for the face particles is not supposed to have a big impact on the IB. However, the IB increased when the resin usage was raised from 10% to 12%. One of the explanations for these results is that raising the resin usage of face particles increased their moisture content that facilitated the heat transfer from the hot-press platens to the core particles and thus facilitated the cure of the adhesive. Separate experiments confirmed that increasing the moisture content of face layer at certain range was indeed able to increase the IB (data not shown).

The effects of the solids content of soy slurry on the IB, MOR and MOE are still poorly understood. It is speculative that the effects have something to do with the penetration of soy in wood particles. For this SF-CA adhesive to work, only the soy on the wood surfaces was available for reacting with CA to form effective adhesives. When the solids content was low, much soy penetrated and remained inside wood cells and was not available for reaction with CA. The viscosity of soy slurry increased along with increasing the solids content. At the certain range, the higher the solids content of soy slurry, the higher the percentage of soy remained on the wood surfaces. However, the soy slurry would not be evenly coated on wood particles, i.e., leaving some particles without any soy on the surfaces, which would consequently reduce the strengths of particleboard panels if the solids content is too high. Results from Fig. 7 and 8 appeared to suggest that around 20% of the total solids content of the soy slurry was optimal for leaving the soy on the surfaces of the wood particles and coating the surfaces with the soy evenly.

Some wood cells were compressed during the hot-pressing and tended to bounce back when the press was open, i.e., the pressure was released. This is commonly called springback in wood composites industry. During the hot-pressing, the platen temperatures used in this study were all above 100 ºC, i.e., above water boiling point. Some moisture inside the particleboard would become steam during the hot-pressing, thus increasing the internal pressure. The internal pressure would disrupt the adhesive bonding among wood particles, which greatly contributed to the springback. The amount of steam generated, i.e., the strength of the internal pressure, was dependent upon the hot-press temperature and time. At a fixed hot-press temperature, the longer the press time, the higher the internal pressure. At a fixed hot-press time, higher the hot-press temperature, the higher the internal pressure. The springback would be reduced and overall strengths of the particleboard would remain high if the adhesive bonding inside the particleboard was strong enough to resist the internal pressure or the sufficient amount of adhesive bonding remained after the internal pressure disappeared. The development of the adhesive bonding required sufficiently long hot-press time and sufficiently high hot-press temperature. Therefore, there were optimum hot-press conditions (temperature and time) in terms of achieving the highest strengths, which was consistent with the results shown in Fig

9-12. Under the optimum hot-press conditions, the adhesive bonding after offsetting its disruption by the internal pressure was the highest.

When soy-coated wood particles without spraying the CA were directly used for making particleboard panels, all panels blew out, i.e., the wood particles did not stick together when the hot press was opened. These experiments indicated that soy flour alone could not be used as an adhesive for forming particleboard. In other words, the CA was an essential component of the adhesive. The CA served as a crosslinking agent that converted soy molecules into highly crosslinked network. The amount of CA had to be sufficient for forming sufficiently high cross-linking density, which explained why the IB and the MOR increased when the amount of CA increased from CA/SF ratios of 1/15 to 1/7. At a fixed amount of the adhesive usage, increase in the amount of the CA accompanied with the decrease in the amount of SF. If the amount of SF was not sufficient to evenly coat wood particles, the strengths of resulting particleboard would decrease. The insufficient amount of SF was speculated to be the reason on why the IB and MOR were lower at the 1/4 CA/SF ratio than at the 1/5 CA/SF ratio.

The storage for the wet soy-coated wood particles might facilitate the wetting and penetration of the soy on the wood particles, thus facilitating the even-coating of the soy on wood particles and consequently the strengths of resulting particleboard panels. This may explain why the IB and MOR increased along with increasing the storage time from 0.5 h to 36 h. It is still poorly understood that the IB, MOR and MOE all decreased when the storage time was raised from 36 h to 48 h.

CONCLUSIONS

The strengths of particleboard increased and then flattened out when the particleboard density was raised from 520 kg/m^3 to 760 kg/m^3. The IB, MOR and MOE all exceeded the industry requirements for the M-2 particleboard under the following conditions: ≥10 wt% of the adhesive usage for the core particles, ≥ 12 wt% of the adhesive usage for the face particles, 10-20% of the total solids content of the soy slurry, 224-245 s of the hot-press time, 180-190 ºC of the hot-press temperature, 1/10-1/4 of the CA/SF weight ratio, and 0.5-36 h of the storage time of the wet soy-coated wood particles. This new method allowed easy preparation of particleboard with this CA-soy adhesive.

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GENERAL CONCLUSIONS

Particleboard is widely used for making bookshelf, furniture, cabinets and many other interior products. At present, particleboard is mainly produced with wood particles and UF resins. The emission of carcinogenic formaldehyde in the production and use of particleboard has generated an urgent need for development of a formaldehyde-free wood adhesive for making particleboard. A formaldehyde-free wood adhesive consisting of soy flour (SF) and a curing agent (CA) CA1000 has been used for commercial production of interior plywood since 2004. This CA-soy adhesive has high viscosity and is difficult to be sprayed onto wood particles with a conventional spraying nozzle. This study developed a new method of using this CA-soy adhesive for making particleboard. This new method involved the coating of wood particles with a dilute soy slurry in water, the drying of the soy-coated wood particles, the spraying of the CA onto the dried soy-coated wood particles, the formation of a particleboard mat with the CA-soy-coated wood particles and the hot-pressing of the mat into particleboard. The high viscosity of the adhesive was no longer an issue with this new method. This new method allowed the strengths of particleboard bonded with this CA-soy adhesive to exceed the industry requirements of M-2 particleboard. The optimal conditions of using this method for making particleboard in terms of enhancing the IB, MOR and MOE were: 760 kg/m^3 of the particleboard density, 11 wt% resin usage for the core particles, 12 wt% resin usage for the face particles, 20 wt% solids content of the soy slurry, 180 °C of the hot-press temperature, 224 s of the hot-press time, 1:7 CA/SF weight ratio, and 36 h of the storage time for the wet soy-coated wood particles.

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