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Soybean-based Green Adhesive for Environment-friendly Furniture Material

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Abstract: Over the last decade, Sick Building Syndrome has become a significant social issue in Korea and many methods have been considered to maintain comfortable indoor air quality. To reduce toxic substances emitted from wood composite products, the source control is an efficient method through the reduction of formaldehyde content by using natural material-based adhesives for composite wood products production. Among alternative materials, soybean protein is considered an appropriate natural material to replace formaldehyde-based resin and many efforts have been made to produce new products, such as soap, shampoo, ink, resin, adhesive and textile through changing the chemical or physical properties of soybean. To process soybeans into these useful products, the beans are dehulled and the oil is removed by crushing at very high pressure or by solvent extraction. For use soybean as an adhesive, it is processed at temperatures below 70°C to preserve the alkaline solubility of the proteins. In addition, soybean-based adhesive is undergone treatment process to improve mechanical properties using urea, urease inhibitor N-(n-butyl) thiophosphoric triamide and sodium dodecyl sulfate. The modified soybean-based adhesive exhibited sufficient mechanical properties to use as an adhesive for composite wood products. This paper is a review article to discuss the possibilities of soybean-based adhesive for environment-friendly furniture materials.

Keywords*: green adhesive, soybean, VOCs, formaldehyde, wood-based panels*

1. Introduction

Wood-based panels are primarily used for industrial applications such as furniture, building materials, and laminate flooring because of their good mechanical characteristics usually high strength and ease of machining and good weathering properties (Suchsland and Woodson 1986). Formaldehyde-based adhesives are currently used for Wood-based panel manufacture. Two major drawbacks of using formaldehyde-based resins are they contain volatile organic compounds harmful to human health and petroleum feed stocks for producing formaldehyde are limited (Li *et al*. 2009).

Formaldehyde-based adhesives such as UF and PF resins dominate the current wood adhesive market. Despite the well-known advantages of such resins, formaldehyde emissions and their nonrenewable nature have become a matter of increasing concern. Therefore, green adhesives from renewable resources and free from formaldehyde are nowadays being developed to replace the UF and PF binders. Unmodified or modified soybean proteins can be used as environmentally friendly and formaldehyde-free substitutes for the traditional synthetic adhesives in wood-based panel manufacturing (Hettiarachchy *et al*. 1995; Mo *et al*. 2001, 2003; Wang *et al*. 2002;

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Cheng *et al.* 2004; Emiliano *et al.* 2010). Soybean protein is an inexpensive and renewable material, and soybean protein adhesives were first developed in the1920s (Kumar *et al.* 2002). Soybean protein adhesives have been used in wood board preparations such as cardboard (Zhong *et al.* 2001), oriented strandboard (Wes- cott *et al.* 2004), wheat straw particleboard (Mo *et al.* 2001, Mo *et al.* 2003) and low density particleboard made from wheat straw and corn pith (Wang *et al.* 2002).

Soybean protein is one of the most important plant protein sources in the world. Its main fraction, b-conglycinin, is a glycoprotein which contains approximately 5% carbohydrate of mainly high-mannose moieties (Kimura *et al.* 1997). Such carbohydrate moieties are similar to the mannosylated receptors on the various cell surfaces of Salmonella, as well as most bacterial species in the family of Enterobacteriaceae, carrying type 1 fimbriae adhesin. Type 1 fimbriae adhesin is widely recognized for its predominant role in the adhesion and colonization of the E. coli and Salmonella species (Beachey *et al.* 1981). Mannose (Firon *et al.* 1983) and mannosylatedglycopeptides (Ashkenazi *et al.* 1991) have the ability to inhibit the adherence of E. coli and Salmonella to their host cells. Recently, Shen, Chen, and Zou (Shen *et al.* 2007) reported that the peptic hydrolysate of b-conglycinin has the ability to maintain the health of mice; the subjects were initially provided with an oral administration of the E. coli O138 infection. The postulated mechanism of effect was an anti-adhesive capacity of the peptide fractions, which were released from soybean b-conglycinin to the common gastrointestinal pathogens. However, there is little information available in the literature about the effect of glycopeptide fractions extracted from soybean on the adhesion of pathogens to their host (Yang *et al.* 2008).

In this study, we discuss the applications of soybean protein for green adhesives and the characteristics of soybean-based adhesive in their application for environment-friendly furniture materials.

2. Soybean-based Adhesive

2.1. Soybean

Soybeans are legumes, the seeds of a low-growing field vine. These vines are ancient in culture; the written record of their domestication in China dates back almost 5000 years (Pen Ts'ao Kong Mu, 2838 B.C.). From that time to until now, soybeans have remained a very important agricultural crop for almost every temperate-climate civilization because of their unusually high content of both triglyceride oil and edible protein.

Furthermore, one of the most striking agricultural developments in the United States in recent times has been the rapid rise of the soybean. While in 1907 there were 50,000 acres of the soybean crop in 1935. This increased to nearly 5,500,000. Seed production amounted to 3,000,000 bushels in 1920 and are increasing to about 40,000,000 in 1935. Remarkable progress has been made in the last few years in developing food and industrial uses for the soybean, using its oil and meal. At present, soybeans are crushed in about 45 oil mills including a few cotton seed oil mills. More than 40% of these mills manufacture soybean food products and soybean flour and more than 75 factories produce various industrial products made from soybeans (Lambuth 1994). More recently, many efforts have been made to produce new products such as soap, shampoo, ink, resin, adhesive and textile through changing the chemical or physical properties of soybean.

Table 1. Commercial forms of soybean proteins (Meyer 1966)

Form	Protein (%)	Fat (%)	Moisture (%)
Flours and grits			
Full-fat	41.0	20.5	-
High-fat	46.0	14.5	6.0
Low-fat	52.5	4.0	6.0
Defatted	53.0	0.6	6.0
Lecithinated	51.0	6.5	7.0
Concentrates	66.2	0.3	6.7
Isolates	92.8	<0.1	4.7

2.2. Preparation of Soybean Adhesive

To process soybeans into these useful products, the beans are dehulled and the oil is removed by crushing the soybeans at a very high pressure or by solvent extraction. If the soybean is intended for adhesive use, it is processed at temperatures below 70°C to preserve the alkaline solubility of the proteins (Prosoy 1948).

The protein content of oil-free soybean meal ranges from about 35 to 55% on a worldwide basis. However, the industrial grades are generally blended to yield a uniform protein content of 44～52%, depending on the source. The other principal constituents of soybean meal are carbohydrates, totaling about 30% and ash, at 5 or 6% (Burnett 1951). The moisture content after processing is quite low, usually less than 10%. Production experience has shown that to perform well as protein glue, adhesive grade “untoasted” soybean meal must be ground into extremely fine flour (Davidson *et al.* 1929). Typically, the dry extracted meal is ground or milled until at least 40%, and preferably 60 - 80%, can pass through a 46-mm (325-mesh) screen. For easier quality control with such fine flours, an alternative specific surface test method is available that determines the average particle size in terms of surface area per gram (Lea *et al.* 1939). For the range of mesh sizes recommended above, the corresponding specific surface values are about 3,000～6,000 cm^2/g.

Soybean flour will wet and swell in plain water but will not disperse, and will thus not yield useful adhesive properties. For this purpose, treatment with a soluble alkaline material is necessary. Almost any organic or inorganic alkali will disperse wetted soybean flour to some degree.

However, soybean wood glues of maximum bonding efficiency require dispersion with some degree of strong alkali such as sodium hydroxide or trisodium phosphate (Brother *et al.* 1940). The effect of this strongly alkaline treatment is to break the internal hydrogen bonds of the coiled protein molecules, literally unfolding them and making their entire complex polar structure available for adhesion to wood. Although essential for adhesion, this alkaline dispersion process exposes the protein structure to gradual destruction by alkaline hydrolysis. Thus, dispersed soybean glue has a limited useful life, slowly losing viscosity and adhesive functionality over a storage period of 6～12 h.

3. Characteristics of Soybean-based Adhesive

3.1. Characteristics of Soybean Protein

Present forms of soybean proteins used as raw materials by the food industry are conveniently classified into three major groups based on protein content. Typical analyses for these products are shown in Table 1. The least refined forms are flours and grits, which have be varying fat contents, particle sizes, textures and degrees of heat treatment (Horan *et al.* 1966). Flours are prepared by grinding soybean flakes to 100 mesh or finer, whereas grits are coarser than 100 mesh. Minimum protein contents of these materials

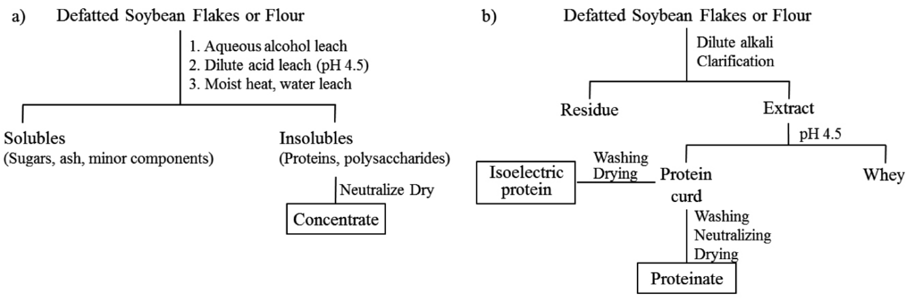

Fig. 1. Flow sheet for a) three methods of commercial preparation of soybean protein concentrates and b) commercial isolation of soybean protein (Meyer 1966).

Table 2. Composition of defatted soy flour (Meyer 1966)

Constituent	Percent
Protein (N × 6.25)	50.5
Carbohydrates	34.2
Fiber	3.2
Ash	5.8
Fat (ether extract)	1.5

range from 40% to 50% depending on the fat content. Defatted flours have approximate compositions as shown in Table 2. Proteins, carbohydrates and ash are the major constituents of defatted flour; the remainder consists of residual lipids and a number of such minor components as saponins, isoflavones and compounds responsible for the typical flavors of raw soybean flour and grits. About one-half of the flour carbohydrates is oligosaccharides-sucrose, stachyose and raffinose, while the other half is made up of polysaccharides, which are insoluble in water or alcohol (Aspinall *et al.* 1967).

Soybean protein concentrates are more refined than flours and grits and contain 70% or more protein on a dry basis. Protein concentrates are prepared from defatted flakes or flour by removing the oligosaccharides, part of the ash and some of the minor components in one of three ways as shown in Fig. 1. The first method involves washing defatted flakes or flour with 60-80% aqueous alcohol (O'Hara *et al.* 1965; Mustakas *et al.* 1962). The proteins and polysaccharides are insoluble in alcohol, while the sugars and other compounds dissolve and are removed. The concentrate is then dried at its natural pH which is near neutrality. A second procedure uses an acid leach at about pH 4.5 to remove the sugars. At this pH the major globulins are at their isoelectric point; both the proteins and polysaccharides are insoluble under these conditions. The leaching step may be carried out at or above room temperature. The wet protein concentrate is then neutralized and dried (Moshy 1964). The third procedure uses moist heat to denature and insolubilize the proteins in the flakes or flour and a water wash is followed to remove the sugars and other minor components (McAnelly 1964). Although concentrates prepared by any of these methods contain 70% or more protein, their physical properties will differ with the method of preparation (Meyer *et al.* 1966). For example, concentrates prepared by acid leaching plus neutralization in the absence of heat treatment will have a higher content of soluble protein than concentrates obtained by heat and alcohol treatments. Concentrates have a reduced flavor level compared to flours and grits because some of the flavor constituents are removed by the concentration processes.

The most refined forms of soybean proteins are

Table 3. Mechanical properties of medium-density wheat straw particleboard as affected by the type of adhesives and chemical treatment of straw (Mo *et al*. 2003)

Sample	TS (MPa)	MOR (MPa)	MOE (MPa)	IB (MPa)
Raw straw				
Methylene diphenyl diisocyanate	11.45	18.1	2281	0.64
Urea Formaldehyde	3.96	6.36	1805	0.11
Soybean Flour	3.36	5.08	1518	0.10
Soybean Protein Isolate	4.21	5.26	1334	0.12
Bleached Straw				
Methylene diphenyl diisocyanate	16.4	26.5	3052	0.87
Urea Formaldehyde	5.74	9.34	1859	0.19
Soybean Flour	4.22	8.04	1851	0.22
Soybean Protein Isolate	6.00	9.60	1601	0.35

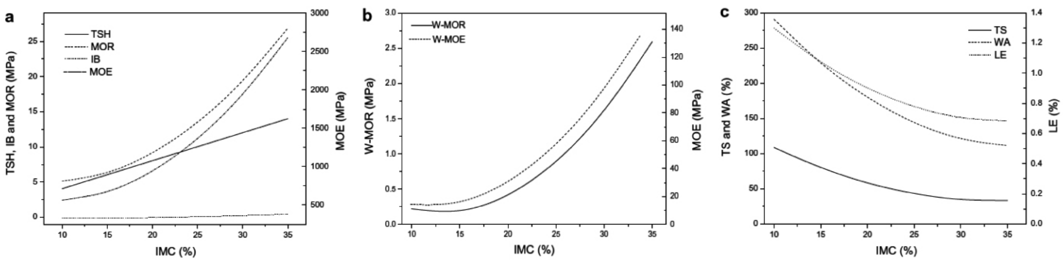

Fig. 2. Initial moisture content of 1% SDS-modified soybean protein adhesive coated wood fiber with (a) tensile strength, internal bonding strength, modulus of rupture and modulus of elasticity; (b) wet modulus of rupture and wet modulus of elasticity after 24 h water soaking; (c) linear expansion, thickness swell and water absorption of wood medium density fiberboard (Li *et al*. 2009).

the isolates, which contain 90% or more protein. They are prepared as shown in Fig. 2 by removing the water-insoluble polysaccharides as well as the oligosaccharides and other low molecular weight components that are separated in making protein concentrates. Defatted flakes or flours, which have received minimum moist heat treatment, are extracted with water plus alkali at a pH of 7 to 8.5. The insoluble residue, which contains the water-insoluble polysaccharides, plus residual protein, is then separated. In the next step the clarified extract containing the bulk of the proteins plus the sugars is adjusted to about pH 4.5. This treatment precipitates the proteins, which are then removed by centrifugation or filtration. The supernatant or filtrate (they shown in Fig. 2) contains the sugars, ash, and minor constituents. The precipitated proteins are then washed and dried to give the isoelectric protein. More commonly, the protein is neutralized before drying. This procedure yields the proteinate form, which has the advantage of being water dispersible, while the isoelectric protein is insoluble in water. Isolates may neutralize more than 95% protein, dry basis, but contain 2~4% ash (Meyer *et al.* 1966) and 3~4 z of minor constituents. The latter are extractable with aqueous alcohol and consist of saponins, phospholipids, sterol glycosides, isoflavones and unidentified compounds (Nash *et al.* 1967).

3.2. Mechanical Properties Improvement of Soybean-based Adhesive

Specifically, phenolic and urea - formaldehyde resins have replaced blood, soybean, and starch glues in all plywood and composite wood panels; resorcinol - formaldehyde resins have replaced casein glues in lumber laminating and millwork applications; and poly (vinyl acetate) and acrylic emulsion glues have replaced virtually all collagen adhesives (animal and fish bone/skin derived) from furniture, musical instruments and general interior wood assembly (Wood Handbook 1987). Limited and specialized applications for protein glues, mainly in combination with synthetic resin polymers, continue to the present.

Most work has been directed toward developing soy protein products with good solubility and adhesive strength for binding pigments in paper coatings and water-based paints. Cone and Brown use alkali to obtain more desirable adhesive properties from soybean flour by controlling the denaturing process of alkali on protein (Cone and Brown 1934). The chemistry and the properties of soy protein related to functional properties in food systems are well documented. Enzymatic and chemical modifications of soy proteins have been used to improve dispersing and emulsifying functional properties. The understanding of the chemistry, selective modification and functional properties of modified soy protein will play a major role in the development of industrial products such as adhesive and binders from soy protein.

The objective of Xiaoqun Mo's research was to characterize the physical properties of medium density straw particleboard as affected by different adhesive types including soybean-based adhesives, UF, MDI as well as chemical treatment of straw. In general, particleboards made from bleached straw showed superior mechanical properties compared with those made from raw straw (Table 3). Although the soybean-based adhesives had poorer mechanical properties than MDI resin, it is environmental friendly and could be used in applications with less stringent requirements for mechanical strength (Mo *et al.* 2003).

The objective of Cheng's research was to improve the mechanical properties and water resistance of wheat straw-soy flour particleboard by chemically modifying soy flour. Urea and urease inhibitor N-(n-butyl) thiophosphorictriamide (nBTPT) were used to modify the proteins. Boric acid and citric acid along with sodium hypophosphite monohydrate were used to modify soy carbohydrates. Sodium hydroxide was used to unfold protein molecules. The combined effect of the chemicals was also studied. Particleboard bonded by urea and high concentrations of nBTPT treated soy flour improved the mechanical properties, but that bonded by boric acid treated soy flour had better water resistance. The adhesive made from soy flour treated with 1.5 M urea, 0.4% n BTPT, 7% citric acid, 4% NaH_2PO_2, 3% boric acid and 1.85% NaOH, produced particleboard with the maximum mechanical strength and water resistance (Cheng *et al.* 2004).

In Xin Li's study, soybean protein was modified with sodium dodecyl sulfate (SDS) as an adhesive for wood fiber medium density fiberboard (MDF) preparation (Li *et al.* 2009). Second-order response surface regression models were used to study the effects and interactions of the initial moisture content (IMC) of coated wood fiber, and the press time (PT), temperature on the mechanical and water soaking properties of MDF. Results showed that the IMC of coated fiber was the dominant influencing factor. The mechanical and soaking properties improved as IMC increased and reached their highest point at an IMC of 35%. Press time and temperature also had a significant effect on the mechanical and

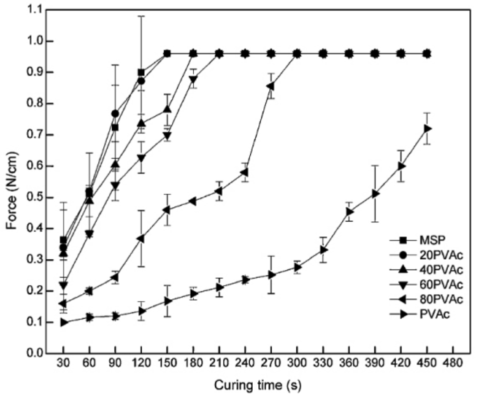

Fig. 3. Effects of PVAc level and curing time on peel strength of MSP on a glass substrate (N/cm) (Guangyan and Xiuzhi 2010).

water soaking properties of MDF. Second-order regression results showed that there were strong relationships between the mechanical and soaking properties of MDF and processing parameters. All mechanical and water soaking properties reached the optimum values at 35% IMC (Fig. 2).

The properties of MDF made using soybean protein adhesive are similar to those of commercial board. The aim of Guangyan Qi's research was to improve the tack strength, curing time, and water resistance by modifying soy protein with a reducing agent and evaluate the resulting adhesive's potential for use on paper labels for glass bottles. The modified soy protein adhesives (MSP) were evaluated for peel adhesion and were compared with adhesives made from soy protein isolate (SPI) and soy flour (SF). Peel strength of the blends (Fig. 3) confirmed that addition of MSP enhanced the peel strength of PVAc between the paper label and glass substrate (Guangyan and Xiuzhi 2010). Polyvinyl acetate (PVAc) is a rubbery synthetic polymer, and PVAc emulsions are widely used as adhesives for wood, paper coating, paint and other industrial coatings. However, the water resistance and gap-filling properties of PVAc are poor. Therefore, commercial PVAc-based latex adhesive was also used for comparison and the compatibility of MSP with PVAc was also studied (Li *et al.* 2009).

4. Conclusions

Recently, concerns about environmental pollution, resource scarcity, and related health issues have pushed scientists to replace synthetic petrochemical polymers, which are used extensively in construction, packaging and labeling industries with bio-based adhesives. Soybean proteins have shown great potential for use as renewable, environmentally friendly adhesives.

Formaldehyde is a suspected human carcinogen that is released from wood-based panels used in home construction including products made with UF resins (e.g., particleboard, hardwood plywood, MDF and paneling). Many consumer products containing formaldehyde-based resins release formaldehyde, leading to consumer dissatisfaction and health-related complaints. These emissions have resulted in a variety of symptoms, the most common of which is irritation to the eyes and upper respiratory tract. However, soybean-based adhesives have been used to replace UF or PF to reduce the emission of formaldehyde.

Not only will this result provide a foundation for the production of environment-friendly products, but a response method for environmental regulations is also reinforced. In addition, the mass production soybean-based adhesive as a natural resource can be used as a substitute to existing adhesives when oil resources are depleted.

Despite all the advantages, however, there are some problems such as the complicated and uneconomical production process. Therefore, research to improve these problems should continue ultimately to produce value-added green adhesives.

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