

AN ABSTRACT OF THE THESIS OF

Suardi Sumadiwangsa for the degree of Doctor of Philosophy in Forest Products presented on December 08 1995.

Title: Viscosity and Bond Quality of Urea-Formaldehyde Adhesive Extended with Acid Modified and Phosphorylated Sago (Metroxylon sp.)

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Abstract approved


Joseph J. Karchesy

It was the goal of this research to determine if sago starch could be used to replace wheat flour as an adhesive extender of Urea-formaldehyde plywood adhesives through either chemical modification of the starch or modification of the adhesive formulation strategy. Guiding principles of the research would be that a useful adhesive would have a viscosity of 1500 cPs and exhibit a curing rate and bond strength comparable to a wheat-flour extended adhesive as measured by the Automated Bonding Evaluation System (ABES) machine.

The research was performed in three stages. In the first stage, the effect of water content, chemical modification, pH, retrogradation, viscosity, water intake and bulk density on the physical properties of sago starch were investigated. Two chemical modifications of sago starch were employed, acid modification with dilute hydrochloric acid and phosphorylation.

In the second stage of experiments, the effect of sago starch content (both natural and modified) on adhesive viscosity was studied where the amount of Urea-formaldehyde resin was held constant.

Data from the first two stages of research were used to formulate adhesive mixtures with 1500 cPs viscosity which were studied for curing rates and maximum bond strength. This viscosity was chosen as one that would fit into current Indonesian plywood production practices. It was found in this stage of the research that all forms of the sago flour studied, natural, acid modified and phosphorylated could be formulated into a Urea-formaldehyde adhesive that gave satisfactory curing rates and maximum bond strengths compared to a wheat flour extended adhesive. For example, a 30% resin solids formulation extended with natural sago, acid modified sago, phosphorylated sago and wheat flour exhibited curing rates of 0.035, 0.030, 0.022, and 0.027 MPa/s respectively. Maximum bond strengths for natural sago, acid modified sago, phosphorylated sago and wheat flour extended adhesives were 9.5, 9.8, 11.1 and 8.9 mPa respectively. Indonesia uses the Japanese Agricultural Standard for certification of its plywood. This standard places paramount importance on bond strength.

Phosphorylated sago may be the best for initial scale up to pilot plant tests since its physical properties were closest to those of wheat flour.

Viscosity and Bond Quality of Urea Formaldehyde
Adhesive Extended with Acid Modified and
Phosphorylated Sago (Metroxylon sp)

By

Suardi Sumadiwangsa

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Suwardi Sumadiwangsa, Author

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this work
is dedicated
to

tintin, dadi
doni, ii, and io

who always fulfil my live
with full of love,
encouragement, and understanding

VISCOSITY AND BOND QUALITY OF
UREA-FORMALDEHYDE ADHESIVES EXTENDED WITH
ACID MODIFIED AND PHOSPHORYLATED
SAGO (METROXYLON SP)

I. INTRODUCTION

Since 1970, both the number and the capacity of Indonesian plywood factories have increased very rapidly. In 1970 there was only one. By 1980 the number had increased to 29; by 1983 there were 79, and by 1986 there was an increase to 108 factories with an actual production equaling roughly 5.8 million cubic meters a year (Anonymous, 1988). By 1991 the real production of the factories had reached 9.95 million cubic meters of plywood per year. The main product has been interior or type II plywood (Anonymous, 1994).

Urea-formaldehyde (UF) is the adhesive of choice for interior plywood. UF resins are relatively cheap, cure rapidly and their clear color does not bleed through to their face veneers. The UF resin is blended with water, hardener, and extender to formulate it into an adhesive mix prior to being spread to the veneer surface. The importance of adding the extender is to control viscosity, to prevent glue line dry-out during panel assembly, and to reduce adhesive cost. Consequently adhesive application (Wheatley, 1991 and Klein, 1980), prepressing and penetration are well controlled. The extenders also possibly improve assembly time and bonding quality of the mixed adhesive (Klein, 1980)

and prevent dry out and under cure (Robertson & Wheatley, 1988). As more extender is added to the adhesive mixture, more water also needs to be added to achieve an optimum viscosity with less UF content. A decrease of UF content makes the adhesive more economical; however, the bonding strength of the adhesive is decreased.

The extender content should be calculated carefully to achieve the most efficient resin glue material and also to satisfy the demands of the adhesive applicator. The mixed adhesive is then applied to the veneer surface. A common method of application is the roller spreader which needs a defined adhesive viscosity to work adequately and to make sure the adhesive is coated homogeneously over the veneer surface.

Depending on the specific application method, different adhesive viscosities are used. Most Indonesian plywood industries use a spreader (Sumadiwangsa, 1992) and a viscosity from 1300 to 1800 centi Poise. Adhesive viscosities will also be adjusted for the species of wood and temperature of the work site. Viscosity adjustments can be achieved by regulating the amount of extender and water added to the adhesive formulation. Typically one kilogram of resin needs approximately 200 grams of extender, 100 grams of water, and 5 grams of hardener. The plywood factories in Indonesia use industrial wheat flour as an extender.

The volume of plywood production in Indonesia is measured in cubic meters (m³) instead of 1000 sq.ft. 3/8" basis as used in the United States of America. Generally, every cubic meter of plywood needs about 84 kgs of UF adhesive, and 16.8 kgs of extender. In 1991, Indonesian plywood factories produced about 10 million cubic meters of plywood (Anonymous, 1994). This production consumed about 177,160 tons of wheat flour that is imported from other countries (Anonymous, 1984). It has been proposed that wheat flour can perhaps be replaced by another starchy material. Like wheat flour, sago flour has a high starch content; therefore, it could be a potential substitute for wheat flour as an extender of UF adhesives. However, the differences in the molecular size and starch content, like gluten, make sago's physical properties unlike wheat flour when it is used to extend UF adhesives.

There are about 200 genera or about 4,000 species of palm trees in the world (Anonymous, 1969). Eight genera of them produce starch from their stems, and five of them grow only in Southeast Asia and Oceania (Ruddle et al., 1978; Johnson, 1977). Metroxylon is the most important genus (Flach, 1983), and it has been exploited on an industrial scale (Knight, 1969).

The most important species in the Metroxylon genus are M. sago and M. rumphii Mart (Flach, 1983). Both can grow up to 10 m in height, and about 40 cm in diameter, and they only flower once during their lifetimes (Flach, 1983). The

sago tree is cut to collect sago flour, usually before flowering occurs or at the juvenile fruiting period.

Practically all parts of the palm tree are useful for native needs, but the most important part is the pith of the stem that can produce sago flour as a native food stuff. The sago flour also is used in the paper and textile industries, as well as in the food industries (Flach, 1983).

Sago flour is water-extracted from the pith. There are three ways to produce sago flour which depend on the size of the industrial user. Often there are some differences in the quality of the various sago flour products.

Sago palm trees mainly grow in Indonesia in addition to Papua New Guinea, Malaysia, Thailand and the Philippines. In Indonesia there are sago molat (Metroxylon sago Roxb.), sago tuni (M.rumphii Mart.), and sago makarano (M. r.var. microcanthum). They grow in Sumatera, Kalimantan Barat (West), Jawa Barat (West), Bali, Sulawesi Selatan (South), Maluku, and Irian Jaya (West Irian). Among them, Irian Jaya and Maluku have the highest concentration of sago trees (Sumarna, 1986; Flach, 1983).

Indonesia has about a 1,114,000 ha of sago palm trees; 1,000,000 ha were estimated from the wild area, and 114,000 ha were from the cultivated area (Flach,1983). If there are 16 to 20 trunks harvested in one ha, and each trunk contains about 850 kgs of pith or 250 kgs of dry flour (Flach, 1984),

so that annually Indonesia is able to produce about 4.5 million tons of dry sago flour from the wild stands. This amount does not include the cultivated stands.

While this field is still new, some research has been done using sago flour as an extender of UF adhesive. Sumadiwangsa (1985) used sago flour exclusively and compared it with wheat flour. Then, Sumadiwangsa (1986) examined the use of sago flour mixed with wheat flour. These experiments on sago flour revealed a poorer quality in bonding strength than that obtained using only wheat flour. Increasing the bonding strength of sago flour probably can be done by modifying sago flour or by adding an enrichment of substitute prior to use as an extender. If the sago flour can be used as an extender of UF adhesive, there will be significant benefits to Indonesia. It would increase work opportunities, land values, native incomes, and in general the spheres of activity would be increased. Indonesian import of wheat flour could then be reduced. In general the dependence on foreign imports will decrease and the values of Indonesian products will increase.

The applied research on sago flour needs to be continued in order to get the most beneficial utilization of sago flour. Use of sago flour as an extender for UF adhesive should be not only practical but also economical.

In this research modified sago flour will be tested for its effectiveness as an extender of UF-adhesives. Meranti (Shorea sp.) veneers will be used as this is the most

commonly used wood in Indonesia. Also, it will be determined in this research if sago flour (modified and unmodified) can be used in a UF adhesive formulation to make satisfactory adhesive bonds.

In these experiments sago starch will be modified by two methods. First, by soaking the sago sample for seven days in 7.5% hydrochloric acid reagent. In the second modifications the sago is soaked one day in the same solution above and then phosphorylated by an experimentally derived mixture of monosodium phosphate and disodium phosphate to achieve appropriate properties as an extender of UF-adhesive as previously mentioned.

Effectiveness of the sago extended adhesives will be evaluated and compared to a standard wheat flour extended adhesive by an automated bonding evaluation system (ABES) developed and improved by Humphrey (1993).

II. LITERATURE REVIEW

2.1 Profile of Indonesian Plywood Factories

Since 1980, the Indonesian government has restricted the export of wood logs as a raw material. According to Governmental Regulation No: 093/Kpts/DJ/I/1980, logs should be converted to lumber or plywood prior to export to another country. The purpose of the regulation is to stimulate the forestry entrepreneur to build mills and manufacture such products as lumber, particleboard, and plywood, and to give added value to the timber resource. In fact, this regulation has been effective in increasing the number of Indonesian wood industries, such as plywood factories (Table 2.1). And then since 1991, Indonesia has become the second highest producer of plywood country after the US (Baldwin, 1995).

In 1983, in Indonesia, there were 79 plywood factories which produced about 2,900,000 cubic meters of plywood. The production increased to 7.4 million cubic meters in 1987, and 9.95 million cubic meters in 1991. Approximately 90 percent of the plywood products were an interior type which used urea-formaldehyde as the resin for the adhesive, and wheat flour as the adhesive extender.

Generally Indonesian plywood factories use an adhesive viscosity of 1300 - 1800 cPoise, and 190 g/m glue spread rate, so that every cubic meter of plywood needs about 84 kg of UF adhesive and 16.8 kg of extender. Therefore, to

produce 9,950,000 cubic meters of plywood, about 835,800 tons of UF adhesive and 177,160 tons of extender are needed.

Table 2.1 Plywood Factories in Indonesia

| Year | Factory # | Prod Cap. | Real Prod* | References |
|------|-----------|-----------|------------|--------------------|
| 1970 | 1 | - | - | Sutigno et al.1983 |
| 1973 | - | - | 0.01 | FAO 1985 |
| 1976 | - | - | 0.22 | FAO 1985 |
| 1979 | - | - | 0.63 | FAO 1985 |
| 1980 | 29 | 1.9 | 1.01 | Sutigno et al 1983 |
| 1981 | 40 | 2.6 | 1.5 | |
| 1982 | 61 | 3.7 | 2.4 | Sutigno et al 1983 |
| 1983 | 79 | 4.5 | 2.9 | Anonymous, 1984 |
| 1984 | 95 | 5.3 | 3.8 | FAO 1985 |
| 1985 | 102 | 6.5 | 5.9 | Anonymous, 1988 |
| 1986 | 108 | 6.8 | 5.8 | Anonymous, 1988 |
| 1987 | - | - | 7.4 | Anonymous, 1988 |
| 1988 | - | - | 7.5 | Anonymous, 1994 |
| 1989 | - | - | 8.9 | Anonymous, 1994 |
| 1990 | - | - | 9.4 | Anonymous, 1994 |
| 1991 | - | - | 10.0 | Anonymous, 1994 |

* in million cubic meters

The importance of UF adhesives and required extenders are increasing due to the increase of new plywood factories (both in number and real production) and also the increasing demand of particleboard factories. Unfortunately, information regarding Indonesian particleboard factories is still not available. However, these factories do increase the need for wheat flour extender but by how much at this time cannot be described.

Extenders are the ingredients added and mixed with resin solutions for several purposes and they have the

ability to slightly increase the quality of adhesion. For urea-formaldehyde resin, there are various types of extender, but the most commonly used are wheat and rye flour, and less commonly, tapioca flour (Perry, 1944). Almost all plywood factories in Indonesia use industrial wheat flour for the extension of UF adhesives. This material is imported from other countries such as Australia and the United States.

2.2 Adhesives and Adhesion

The quality of wood adhesion (Scheberger, 1985) depends upon wood properties, bond formation, bond properties, bond failure and gluing conditions. Each of these variables depends on many disciplines such as chemistry, physics, rheology, material behavior, surface science and thermodynamics (Scheberger, 1985) which affect the final quality of adhesion. There are two kinds of force or work (Huntsberger, 1970) in adhesion: **a) adhesion**, the force of attraction or bond energy, and **b) adhesive performance**, the force to separate the bond. These two kinds of force are expected to have the same value; however, they sometimes differ by several orders of magnitude (Huntsberger, 1970)

One of the criteria for predicting the bonding strength of an adhesive is wettability, the ability of a glue liquid to make intimate contact with a wood surface (Wellons, 1980). The relationship between bonding strength and wettability is significant, especially when bonding urea-formaldehyde resin to wood (Wellons, 1980).

Adhesives cure at very broad range of speeds, from seconds to months, (Marra,1992) depending on their composition and the conditions under which they are cured. Curing rate determines the rate of production and controls the magnitude of adhesive mobility prior to gelation. Basically, curing rate (Marra,1992) is "a built-in characteristic of the adhesive", however, it can be controlled by:

- 1) the use of additives, catalysts and solvents
- 2) wood conditions: temperature, moisture content, surface roughness, wood species, and thickness
- 3) the age of the adhesive
- 4) the spread rate
- 5) the glue line temperature and the rate of increasing temperature.

During curing, adhesive will flow throughout the bond line, transfer on the unspread surface, penetrate to the wood pores and capillaries, wet the wood surface and solidify (cure) throughout the adhesion layer (Marra,1992).

Adhesive should be used at a proper curing rate. If the curing rate is too fast then insufficient penetration and wetting will occur; if too slow, complete wetting will occur but possibly with over penetration (Marra,1992).

2.2.1 Veneer

The critical wood variables that can alter the adhesive bond strength are as follows:

- moisture content

- structure (earlywood vs. latewood, density, porosity)
- chemical components (cellulose, lignin, extractives)
- surface quality (smoothness, texture, energy, and fiber direction)

The moisture of the veneer and of the adhesive mixture can control both the penetration depth and the curing time. Too much water in the veneer (Zavala, 1985, and Resch, 1970) causes "wash out", "blows" (steam blister), and resin penetration too deep into wood pores.

Differences in wood density between adherents can also cause problems; for example, gluing latewood to latewood may cause lower shear strength than earlywood to earlywood (Kozlik, 1974). Also, in latewood to earlywood bonds it can sometimes be difficult to get the correct amount of adhesive penetration into each due to their different abilities to absorb the adhesive. In veneer production, summer wood is likely to peel between cells or in the outer part of the cell wall. As a result, the veneer surface is lignin rich. On the contrary, springwood peels across the cell wall, so that the veneer surface is hemicellulose rich.

The major components of wood are cellulose (65-75 %), lignin (20-30%), and extractives (0-10%). Cellulose is a polar molecule with a high surface energy that bonds effectively. Lignin is less polar; thus the surface has lower energy and bonds less effectively.

Some of the extractives are polar molecules such as sugar; but others are non-polar, such as oleo resins and

fatty acids. Fatty acids contribute to a downgrading of the bonding ability of wood. The adhesive cannot wet the wood surface because the surface is blocked by the non-polar fatty acids. The bonding strength between sapwood and heartwood can also be different due to pH, which is related to the extractive content of mildly acidic phenols which are found mostly in the heartwood segment. During high temperature drying, the extractives may also melt and migrate to the veneer surface (Resch,1970). The veneer surface then becomes less polar and has less wettability for the water borne wood adhesive mixtures.

In view of the potential for dehydration reactions of cellulose hydroxyl groups at high temperatures, overheated drying develops inactivated veneer surfaces which are less polar. This happens at temperatures above about 150° C. At this temperature or above, hydroxyl groups (Resch,1970) form less-polar ether linkages. Less polarity negatively effects the adhesion ability.

The size and consequence of surface inactivation depends on time, temperature, moisture content, air/steam velocity (Kozlik,1974), and wood species. Generally, veneer drying at 205° C. for one or two minutes is safe. When dried at 150° C. for 20 minutes, however, the veneer becomes surface inactive.

2.2.2 Adhesives

Wood can be adhered by many kinds of glues or adhesives. Plywood veneer, however, basically is glued with

two kinds of adhesives: exterior or structural adhesives, such as phenol-formaldehyde (PF) and phenol-resorcinol-formaldehyde (PRF), and interior or decorative adhesive such as urea-formaldehyde (UF) or melamine-urea-formaldehyde (Lambuth, 1977). These kinds of adhesives (resins) are solid substances (mostly powder or flake) and are difficult to apply uniformly to plywood veneers. Usually they are dissolved with a solvent prior to being applied to the plywood veneers (Sellers, 1985).

Solvents, such as water or alcohol, can be used to dissolve these resins. Such a liquid mixture is able to move and spread easily and homogeneously on the surface of plywood veneers. Theoretically, the straight (neat) resin (resin in its solution) is the highest in the bonding strength for adhesion properties. For some reasons cited below, the application of the straight (neat) resin may, on occasion, not have sufficient properties to spread on the veneer surface well. There are also economic factors which must be taken into account.

When preparing an adhesive, a compromise has to be struck between viscosity and resin content. It is hard to achieve an adequate viscosity for the spreader while the resin content is also appropriate to reach the desired adhesion properties (Sellers, 1988). If the viscosity is appropriate for the spreader the resin content is usually too high, so it is not economical. On the contrary, if the resin content is sufficient from the point of view of

adhesion properties, the viscosity is under the spreader ability, so it fails in the spreading process.

Second, the straight resin does not have "a body", so "dry out" of the glue-line and inappropriate penetration into the wood can not be controlled. Neglecting the "dry out" of the glue-line causes delaminations (Robertson & Robertson, 1977).

Third, the straight resin changes its viscosity easily, especially in storage prior to being spread on the plywood veneer surfaces (Robertson & Robertson, 1977).

All of these glue problems can be solved by adding an extender, or a filler, or sometimes both materials. UF resins, for example, are often mixed with wheat flour (extender) and pecan or walnut shell flour (filler) (Klein, 1980).

2.2.3 Extender

Often the term extender and filler are interchangeable (Klein, 1980; Robertson & Robertson 1977; and Sellers 1985). Both materials have the same functions as follows (Sellers, 1985):

- a. tack consistency
- b. better adhesion on dense woods
- c. greater assembly time tolerance to "dry out"
- d. better resistance to over penetration
- e. faster and stronger pre-press bonds.

The difference between fillers and extenders are in their abilities to adhere to wood components. An extender,

in this case, has little adhesion properties, whereas a filler does not have any ability to adhere to wood components (Klein, 1980; Robertson & Robertson, 1977; and Sellers, 1985).

Robertson & Robertson (1977) classified extenders as glutinous extenders, such as wheat flour, and non glutinous ones. Moreover, non glutinous extenders are also divided into cereal products such as rye-, sorghum-, and soy- flour, and non-cereal products like blood flour.

Wheat flour is the main extender substance (Sellers, 1985; Robertson & Robertson, 1977). This protein starch extender increases glue-line quality and glue properties caused by its components, such as starch and protein (Robertson & Robertson, 1977).

2.2.4 Extender Characteristics

The reason for using extenders on different types of plywood are generally the same. In certain situations, however, there are differences between the structural (softwood) and interior (hardwood) plywood (Sellers, 1985). Structural plywood consumes a secondary extender, while interior plywood formulations use a primary extender.

The quantity of extender solids used in the hardwood plywood industry is often proportional to the quantity of UF resin solid consumed. The glue mixture consists of water, resin, extender, and a catalyst. This mixture should be able to flow to the orifices of curtain coaters and spray

applicators, and remain stable in its viscosity during storage prior to being spread on the veneer surfaces (Sellers, 1985).

Klein (1980) provides a list of commonly used extenders as followings:

Wheat flours

wheat starch, protein content 0 - 2 %

soft wheat flour, protein content 7 - 11 %

durum wheat flour, protein content 10 - 14 %

hard wheat flour, protein content 13 - 18 %

Dried animal blood (beef, hog, sheep)

Soluble blood

Low soluble blood

High viscosity low soluble (HVLS) blood

Feed grade blood

Sorghum (Milo) flour

Rye flour

Potato starch

Corn starch

Corn flour

Robertson & Wheatley (1988) stated that the best extender is Medium Gluten Soft Winter Wheat. The gluten content is about nine percent. Higher contents than that will reduce flowability, need more wash water, and shares (contributes) to blow. Lower gluten contents lead to reduced

tackiness. Ash content should be low to reduce abrasiveness. Viscosity and moisture content of an extender should be consistent.

2.3 Sago Palm Trees and Sago Flour

It is believed that there are 200 genera, which consists of 4,000 species of palms in the world. The kingdom of the palms, mainly tropical plants, are divided into two hemispheres; the Western and Eastern. In the Western hemisphere there are about 1,250 species. Brazil has about 500, and Columbia has more than 250 species (Anonymous, 1969). The rest (about 2,750 species) grow in the Eastern hemisphere (Asia and Oceania).

2.3.1 Carbohydrate Producing Palms

Among the palms, there are eight genera that have stems able to produce carbohydrates, and five of them grow only in Southeast Asia and Oceania (Ruddle et al., 1978; Johnson, 1977). The most important genus (Flach, 1983), Metroxylon, has been exploited on an industrial scale (Knight, 1969).

The sago palms mainly grow in a longitude of 90° to 180° east, and in a latitude of 10° north to 10° south. They grow at an altitude of up to 700 meters above sea level (Flach, 1983). Their distribution is mainly on the Irian island (Papua), which is believed to be the gene center (Flach, 1977 in Ulijaszek, 1984). The countries where the sago palms stand abundant are shown in Table 2.2. Here it

can be seen that Indonesia has the largest area of the sago palm. The distribution of sago palm trees in Indonesia are shown in Figure 2.1.

Yearly, every hectare of natural stands is able to produce 2.5 to 5 tons of dry sago flour. Meanwhile, a hectare of semicultivated and well cultivated stand is qualified (Flach, 1983) to produce 10 and 25 tons of dry sago flour, respectively (Table 2.3).

Sago palms are members of the Lepidocaryoid sub-family of the Palmaeae (Arecaceae). The species that are economically most important in the Metroxylon genus are M. sagus ROTTB. and M. rumphii MART (Flach, 1983). These sago palms can grow up to 10 m in height, and about 40 cm in diameter, with large and pinnate leaves. They are a suckering perennial; they only flower once during the life span and then die (Flach, 1983).

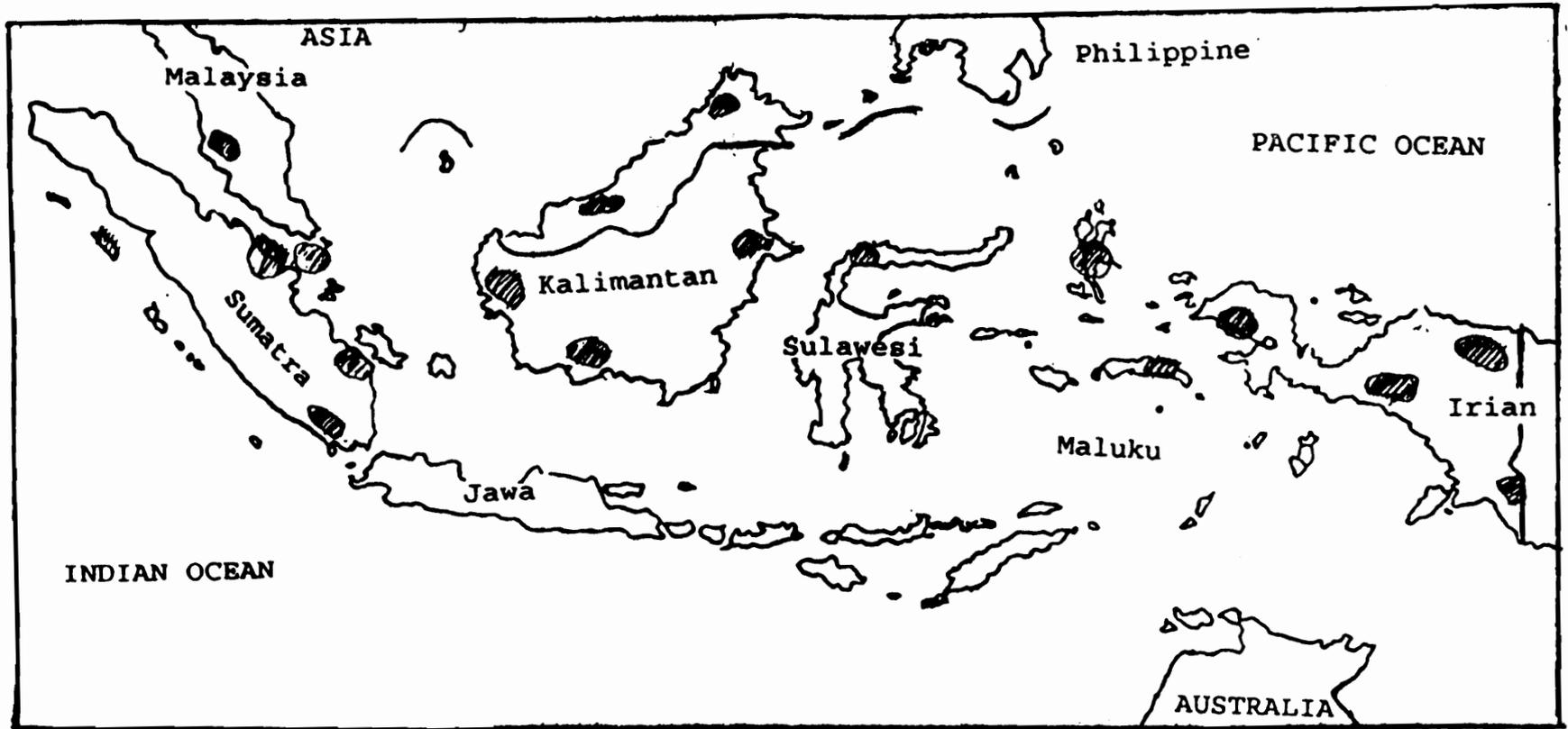


Figure 1.1. Distribution of sago palm trees in Indonesia

Table 2.2 Estimated Area of Sago Palm Trees

| | Natural, ha | Cultivated, ha |
|-------------------------|------------------|----------------|
| Sepik Province | 500 000 | 5 000 |
| Gulf Province | 400 000 | 5 000 |
| Other provinces | 100 000 | 10 000 |
| PAPUA NEW GUINEA | 1 000 | 20 000 |
| Irian Barat | 980 000 | 14 000 |
| Cendrawasih | 100 000 | 2 000 |
| Lake plain | 400 000 | - |
| South Irian | 350 000 | 2 000 |
| Other districts | 130 000 | 10 000 |
| Maluku | 20 000 | 10 000 |
| Sulawesi | - | 10 000 |
| Kalimantan | - | 20 000 |
| Sumatera | - | 30 000 |
| Riow islands | - | 20 000 |
| Mentawai islands | - | 10 000 |
| INDONESIA | 1 000 000 | 114 000 |
| Sabah | - | 10 000 |
| Serawak | - | 20 000 |
| West Malaysia | - | 3 000 |
| MALAYSIA | - | 33 000 |
| THAILAND | - | 5 000 |
| PHILIPPINES | - | 5 000 |
| PACIFIC ISLANDS | - | 10 000 |
| TOTAL | 2 000 000 | 200 000 |

Source: Flach, 1983

Table 2.3 Estimation of Yearly Sago Flour Production

| Sago Stands | tree # /ha | yield/tree (kg) | yield/ha (ton) | Location |
|-----------------|---------------|--------------------|-------------------|---------------------|
| Natural | 16-20 25 | 150 100 | 2.4-3 2.5 | Papua New Guinea |
| Semi Cultivated | 50 30-36 | 200 - | 10 7.8-10.6 | Siberut island |
| Well Cultivated | 136-139 | >175 | >25 | West Malaysia |

Source: Flach (1983)

When there is no cutting, the flowers will produce up to 900,000 seeds. The life cycle fluctuates between 8 to 17 years (Flach, 1983). The multiplications are either by generative (seed) or vegetative (sucker). However, the most common regeneration happens, either natural or plantation, by sucker formation (Flach, 1983). The sago tree is cut and processed to collect the sago flour before the flowering occurs or at the young fruiting stage.

| | | |
|----------|------------------|------------------------------|
| | Trunk cortex: | fuel, construction |
| | Leaflets: | Thatching, house building |
| SAGOPALM | PITH: Processing | STARCH, and Fiber |
| | Rachis: | house building |
| | Rasped: | animal feed |

Figure 2.2 The Uses of the Sago Palm Trees

Almost all parts of the palm trees can be used for some kinds of native needs. The leaflets can be used for thatching and house building. Pith also can be used for house building, fuel, and animal feed (Flach, 1983). The uses of the sago palm tree components are quite diverse (Figure 2.2).

The most important utilization of the pith, however, is for the production of sago flour. The sago flour is mainly used by natives as a food stuff. It is also used in paper and by textile industries as a coating and sizing agent and in the food processing industry (e.g., dextrose and glucose) as an extender or viscosity control agent (Flach, 1983).

Sago flour is water extracted from the pith of the sago palm stem. Technically, however, there are several ways to produce it, such as at a native (village) level and in small factories, which employ simple mechanical equipment. Unfortunately, there is no information about the efficiency of these levels of production. Obviously, there are some differences in the quality, because the quality of water used for extraction determines the quality of the sago flour product (Flach, 1983). High quality sago needs high quality water. Poor quality water causes colored sago flour (greyish/brown) or it has high levels of contaminants.

In natural areas, the trees are cut at the young fruit stage about 1 m above ground level. In commercial planting, the trees are cut before the initial flowering, and they are cut at ground level. Before cutting, the stem is bored to sample the starch content of the pith. There are two ways to determine the starch content in situ. One is drying the pith sample on the axe surface. Another method is by chewing the pith sample. The starch content may be too low or suitable. If the starch content is too low, the hole is plugged (Flach, 1983) with mud. This technique allows the young tree to cure and continue to grow to increase the starch content. The tree will be evaluated at a later time.

When the starch content is sufficient, it is the time for the tree to be cut. The fallen tree is processed to get sago flour by the following steps (Flach, 1983). First the trunk is cut into lengths of 1 to 1.2 m. and split off into 4 or 6 parts. Then they are rasped, such as by a nailed board. After that, 'fibers' are kneaded and sieved by hand on flowing water. The sago flour will settle. Finally the wet sago flour is dried by sunlight.

Table 2.4 Composition of Sago Palm Trees
a. From Serawak and Irian Barat (kg)

| | Serawak | | Irian Barat | |
|---------|---------|-----|-------------|-----|
| | Fresh | Dry | Fresh | Dry |
| Trunk | 875 | 351 | 1250 | - |
| Cortex | 225 | 48 | 400 | - |
| Pith | 650 | 303 | 850 | 425 |
| Starch | - | 164 | - | 250 |
| Water | 347 | - | 425 | - |
| Residue | - | 139 | - | 175 |

Source: Flach (1983)

Table 2.4 Composition of Sago Palm Trees
b. According to the Other References

| | Colon (1958) | Vegter (1983) | Toyo Menka Kaisha (1972) |
|---------|-----------------|------------------|-----------------------------|
| Trunk | 1250 | 875 | 800 |
| Cortex | 400 | 225 | 157 |
| Pith | 850 | 650 | 643 |
| Starch | 250 | 164 | 138 |
| Water | 425 | 347 | - |
| Residue | 175 | 139 | - |

Source: Flach (1983)

Building a sago flour factory has some advantages and disadvantages. With the factory, the opportunity cost of the palm trees can be used and land values increased, as well as native income and the sphere of activity to the native people. The disadvantage of factories, on the other hand, are its environmental consequences in the vicinity and in the forest area due to over production of feedstock, if any, and pollution from waste water and some residues. The utilization of the fibrous residue would reduce the possibility of environmental pollution. The possibilities to use it are such as a feedstock in the fiberboard industry, mushroom media, and duck feed.

The trunk of the palm tree can be divided into two parts, which include the cortex (bark) and the pith (inner trunk). Their properties are obviously very different. The cortex is much harder and stronger than the pith. Flach (1983) noted that based on the total fresh weight, the weight of the sago palm trunks vary from 800 - 1,800 kg. The cortex constitutes 25.7-32.0% and the pith 68.0-74.2% of the stem. Furthermore, based on fresh total trunk weight, the pith contains 18.7 - 20.0% starch, 34.0 - 39.7% moisture, and 14.0 - 15.9% other materials such as fiber and ash. The trunk composition can be seen in Table 2.4.

The pith of the sago palm trees is almost entirely different from the wood of the woody plant species such as in its anatomy, structure, as well as in chemical and mechanical properties. The vessels are randomly spread. The

largest diameter of the vessel is about 1.4 mm. The fiber bundles can be seen with the naked eye. The size of starch granules is between 40 - 50 μm , and the size of the intercellular spaces between the pith cell (Flach, 1983) may be up to 200 μm .

2.3.2 Sago Palms in Indonesia

In Indonesia there are sago molat (Metroxylon sagus ROTTB.), sago tuni (M. rumphii MART.), sago makarano (M.r. var. longispinum), sago ihur (M.r. var. silvestre), and sago rotan (M.r. var. microcanthum). They grow in Sumatra, Kalimantan Barat (West), Jawa Barat (West), Bali, Sulawesi Selatan (South), Maluku, and Irian Jaya (West Irian). Among them Irian Jaya and Maluku have the highest concentration for sago trees (Sumarna, 1986).

Flach (1983) has calculated that yearly each hectare of natural, semi cultivated, and well cultivated palm trees are able to produce 2.4 to 3, 10, and 25 tons of sago flour, respectively. When the area of sago palm trees are calculated to estimate the yield, Indonesia has the potency to produce, at least, 2.7 and 2.0 million tons of sago flour from natural trees and cultivated trees, respectively. Therefore Indonesia has a total capacity to produce about 4.7 million tons of sago flour (Table 2.5).

Table 2.5 Estimated Production of Indonesian Sago Flour (ton)

| | Natural | Cultivated |
|------------------|-----------|------------|
| Irian Barat | 2 646 000 | 245 000 |
| Cendrawasih | 270 000 | 35 000 |
| Lake plain | 1080 000 | - |
| South Irian | 945 000 | 35 000 |
| Other districts | 351 000 | 175 000 |
| Maluku | 54 000 | 175 000 |
| Sulawesi | - | 175 000 |
| Kalimantan | - | 350 000 |
| Sumatera | - | 525 000 |
| Riow islands | - | 350 000 |
| Mentawai islands | - | 175 000 |
| TOTAL | 2 700 000 | 1 995 000 |

Calculated from Flach (1983)

In Siberut island, west of Sumatera island, in every ha there are 30 - 36 trunks of palm trees harvested yearly. The product is about 7800 kg to 10600 kg of dry starch (Flach, 1983) per ha per year.

2.3.3 Sago Flour Utilization

Sago palm is a multiuse tree. Almost all parts of the tree can be used for human purposes. Baay (in Flach, 1983) has devised a use for each part of the tree, as seen in Figure 2.2. The main importance of the tree, however, is pith that produces sago flour.

There are two kinds of materials in the pith. The first one is sago flour, which is mainly used as a native food stuff. Another one is residue, cellulose fiber, that

usually is thrown away. It is possible to use the residue for animal feed, fiberboard making, or as media of mushroom cultivation.

Even though there are many uses for sago flour (Figure 2.3), this paper will concentrate on its use as an extender for urea-formaldehyde adhesive. The basic reason that sago flour can be used as an extender of adhesive is that it contains starch as the main chemical component.

The starch of sago flour is assumed to be able to increase the viscosity of the UF resin. Therefore, sago flour can be used as a viscosity control agent. Industrial wheat flour (IWF) is the carbohydrate that has been commonly used for this purpose. Usually IWF is added up to 50% based on the resin weight (Sellers, 1985). The greater the amount of IWF added to the adhesive, the lower its bonding strength.

Like wheat flour, sago flour has a high carbohydrate content. Therefore, it could be a potential industrial wheat flour substitute as an extender for UF adhesive.

| | | |
|-------------|---------------------|-------------------------------|
| | food | bread |
| | feed | duck-feed |
| | paper industry | |
| SAGO STARCH | starch-derivatives: | oxidized, esters and ether |
| | modified starch: | dextrines, pyro-dextrines |
| | textile industry | |
| | plastic filler | |
| | dextrose, glucose: | ethanol |

Figure 2.3 The Uses of Sago Starch

2.3.4 Research on Sago Flour as an Extender

Sumadiwangsa (1985) first explored the use of sago flour as an extender for UF adhesive in plywood manufacturing. The variables that were explored included: sago origins, veneer species, resin manufacturers, and extender contents. Among those variables, only the extender content was found to influence the plywood bonding strength.

Compared to wheat flour, sago flour has poorer quality for use as an extender. Probably the difference is caused by the minor chemical components in sago flour. An important component that could effect the extender quality is protein. Sago flour has a lower protein content than wheat flour has.

In an additional study, Sumadiwangsa (1986) further explored the use of sago flour that was blended with wheat flour in several ratios. The variables that were observed in this experiment were extender ratio factors and extender content factors. Ratio factors (wheat/sago) consisted of 4 levels that were 10/0; 8/2; 6/4; and 4/6. Extender contents consisted of 4 levels that were 15%; 20%; 25%; and 30% based on resin weight. This experiment showed that the ratio of extender does not influence the bonding strength of plywood. However, the extender content influenced the bonding strength of plywood significantly.

In a third study (Sumadiwangsa, et al. 1987) studied was the use of sago flour in a blend with the tuber flour of iles-iles (Amorphophallus oncophyllus). The Amorphophallus flour contains mannan that is used as an adhesive for wood or paper. This experiment showed the ratio sago/iles-iles at 7/3 is almost equal to wheat flour as an extender of UF on plywood manufacture.

2.4 Starch: Definition and Properties

Starch is a white powdery polymeric carbohydrate substance (a polysaccharide) produced especially by the edible plants. It is thus a renewable substance from a utilization perspective. Usually, it is (Anonymous, 1964) deposited in the seeds (corn, wheat, rice), in roots or tubers (tapioca, potato, sweet potato), or sometimes in the stem pith (sago). Naturally starch is used by a tree as a reserve food (Anonymous, 1964). Starch is a meaningful

material. It is vital not only for food stuff or food industries, but also for non food industries such as in paper, textile, and adhesive manufacture.

The basic chemical elements of starch are C, H, and O. Since the amount of H atoms are always double of that O atoms, the former scientists assume that the starch molecule is a carbohydrate $[C_n(H_2O)_m]$. Later enough evidence was accumulated to conclude that the linkage between O and H atoms is not H_2O (water) molecules, but hydroxyl (OH) groups.

By simple calculation, the empirical formula of starch is $C_{6n}H_{2(5n+1)}O_{(5n+1)}$ [n is the number of saccharide or -D-glucopyranose units]. This empirical formula also can be used for another carbohydrate like cellulose. The monomer of cellulose also is glucose but with 1→4 β linkage rather than linkage 1→4 α like in starch.

Without magnification (Anonymous,1964) there is no difference among the dry starches. Under a microscope the size and shape of the starch granules are specific to the botanical variety. Hence, the microscope is often used as a method to determine the origin of starch (Anonymous,1964).

The main components of starch are amylose, amylopectin, water, protein, fatty acid, and ash. Amylose and amylopectin with iodine (Anonymous,1964) give blue complexes and a red or plum color, respectively. With the higher (amyl) alcohols or fatty acids (Anonymous,1964) amylose produces insoluble complexes, whereas the amylopectin remain

in solution. Amylose is a linear chain of dextrose polymers, while amylopectin is a branched chain (Anonymous, 1964).

Starch granules are insoluble in cold water (Wurzburg, 1986). They vary in size and shape, depending upon their source. Rice starch is polygonal in shape, 3-8 μm in diameter. Corn starch is generally polygonal, but some round granules with 5 to 25 μm in diameter are known. Tapioca starch is rounded, truncated at one end, 5-35 μm in diameter. Wheat starch can have flat, round, or elliptical granules which tend to be in clusters (2-20) and (20-35 μm). Potato starch has the largest (15-200 μm) granules of any commercial starch. They are oval or egg-shaped (Wurzburg, 1986). The properties of wheat and sago starch are shown in Table 6.

Table 2.6 Properties of Wheat and Sago Starches

| Starch type | Wheat | Sago |
|---------------------------------------|-------------------|--------------------------|
| Granule size (μm) | 2-10 and 20-35 | 20-60 |
| Granule shape | Round, elliptical | Egg like, truncated |
| Polarized light | Black cross | Irregular black cross |
| Amylose/Amylopectin | 25/75 | 27/73 |
| Gelatinization ($^{\circ}\text{C}$) | 52-64 | 60-72 |
| Lipid (%) | 1.0 | Very low |
| Paste clarity | Opaque | Translucent |
| Paste texture | Short, heavy body | Long, stringy heavy body |
| Viscosity | Medium/low | Medium/high |

Source: Knight, 1969.

2.5 Starch Modifications

Starch can be used either in a natural or modified form. Often, to fulfill specific industry requirements, the starch generally must be modified, converted or derivatized (Ruttenberg, et al. 1984). Modification can be achieved by genetic control (corn, wheat, and rice) or by physical, mechanical, or chemical actions (Wurzburg, 1986). Sago starch, however, has not yet been genetically manipulated. The modification of sago starch should be done by any of the three external actions mentioned above.

Modification by chemical action may include just the reduction of the length or the size of the molecule (Hydrolysis, and dextrinization) (Ruttenberg, et al. 1984) or by changing some of the chemical structure of

D-glucopyranosyl units. Hydrolysis and dextrinization apparently are the first steps to prepare more suitable material for the next modifications. Generally, modifications involve molecular scission, molecular rearrangement, oxidation, esterification, or etherification (Wurzburg, 1986). The modification changes the chemical or physical properties of starch, making them more useful in a myriad of industrial applications (Wurzburg, 1986).

Modification of starch is employed to control gelatinization and cooking characteristics of natural starch, to decrease retrogradation, to increase the water holding capacity, to enhance hydrophilic character, to impart hydrophobic properties, or to introduce ionic

substitute (Ruttenberg, et al. 1984). Modification is a significant factor in the continuity and the increased use of starch to furnish thickening, gelling, binding, adhesive, and film forming functionality (Ruttenberg, et al. 1984).

There are many methods to modify starch such as hydrolysis, oxidation, pyrolysis, acetylation, hydroxyalkylation, hydroxypropylation, phosphorylation, grafting, and cross linking.

2.5.1 Acid Modification

The modification of starch by an acid was done first by Kirchoff in 1811 (Wurzberg,1987), who used acid on a dispersed starch. Later Naegeli used cold aqueous hydrochloric acid on granular starch for several weeks. In 1886, Lintner first made what is now called Lintner Starch. This is a product of potato starch which is treated by a mineral acid for several days. Belmas and also Duryea (Wurzberg,1987) treated starch at relative high temperature (below gelatinization temperature), with a weaker acid, and for shorter reaction times than Naegeli or Lintner.

Acid will break the glucosidic linkage on the starch which will make the chain length shorter (depolymerization). Swanson and Cori in 1948 and Wolfram in 1963 (Wurzberg,1987) stated that the 1 → 4 linkages are more easy to hydrolyze than the 1 → 6 linkages. In the first stage of hydrolysis, the acids attack the amorphous region of an amylopectin (Wurzberg,1987) followed by the crystalline region of both an amylose and an amylopectin. The mineral acids commonly

used for acid modification are hydrochloric acid, and sulfuric acid (Wurzburg,1987) .

Acid modified starch (AMS) has lower hot paste viscosity (or higher hot paste fluidity), smaller molecular weight, lower intrinsic viscosity, higher in the alkali number, and higher hot water solubility. The Iodine Affinity Number of AMS is uncertain because it depends on the kind of starch (Wurzburg,1987). Microscopically, at room temperature, the granule of AMS is likely to be the same as the parent starch. But when heated in water it will swell less, and the gelatinization temperature is higher than that of unmodified starch.

Due to its low viscosity, AMS needs more quantity than the parent starch to get the same viscosity in a mixture. The behavior of AMS hot paste is less thixotropic, more Newtonian, and is a more clear fluid (Wurzburg,1987) .

2.5.2 Phosphate Modification

The product of phosphate modification is a starch phosphate ester (SPE). There are three kinds of SPE: monoester (mono starch phosphate ester), diester (distarch phosphate ester), and triesters (tristarch phosphate ester) (Figure 2.4). Diester and triesters are the cross linked types of starch phosphates.

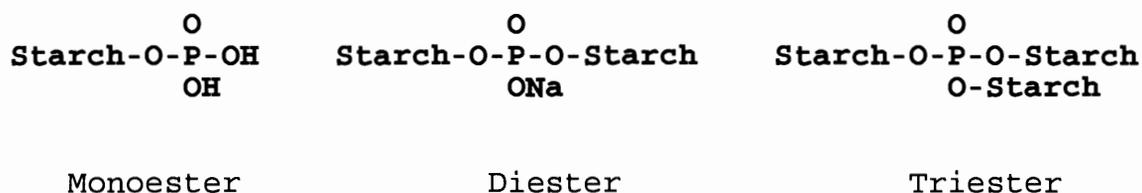


Figure 2.4 Starch Phosphates

Phosphorylation of starch usually employs some inorganic phosphate salts or specific organic reagents. The phosphate salts are the water soluble type like ortho-, pyro-, meta-, or tripolyphosphate. Mixtures of monosodium hydrogen phosphate and disodium hydrogen phosphate are suitable for making mono esters of starch phosphate with up to 0.2 D.S. (Wurzburg, 1987). The other phosphates desirable to prepare monophosphates are sodium hexametaphosphate, tetrasodium pyrophosphate (TSP), a mixture of TSP with phosphoric acid, orthophosphate, or sodium tripolyphosphate. Instead of sodium, other metallic salts of potassium, lithium, or ammonium phosphate are employed in phosphorylation.

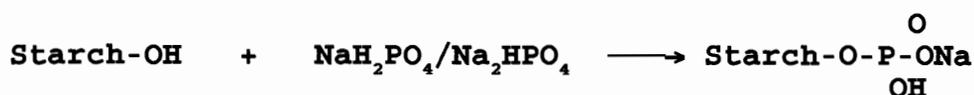


Figure 2.5 Reaction of starch and sodium ortho phosphate

To impregnate with phosphate salts, starch is suspended in the salt solution or starch slurry, which is prepared by adding a certain amount of salt. Using orthophosphate the mixture acidity is controlled at 4 to 6.5 pH. This mixture

is mixed for 10 to 30 minutes and then filtered and dried. The heat of reaction is between 140 to 160° C. for two hours or longer (Wurzburg, 1987). The phosphorylation pH is very critical. A lower pH creates more hydrolysis on the glycosidic linkages, so the products have lower viscosity. On the contrary, too high pH will increase diester phosphate or increase formation of cross linking products. Urea (water soluble organic amides) insignificantly increases the reaction between phosphate salts and starch which produce high viscosity and colorless products. SPE dispersion is very stable; it is comparable with gelatin, vegetable gums, polyvinylalcohol, and polyacrylates. Among the modification starch, SPE has superior properties on freeze-thaw stability.

SPEs are consumed on wet-end additives in paper making. At pH 5.5 about 0.5 to 2.0 % SPE is almost completely retained in the fiber pulp. Choosing SPEs on the wet end can improve retention and strength. Starch phosphate blended with a chelate solution containing alum, tartaric acid and ammonia improves coating composition, better storage ability, water resistance and printability.

A starch phosphate blend with neoprene rubber latex has glue strength, on wood pieces, higher than latex alone. Starch monophosphate is useful in warp sizing, textile printing, and stiffening. They are also efficient, easy to remove and precipitated from the waste water.

2.5.3 Other Modifications

Sodium or calcium hypochlorite are the oxidation agents which are mainly used to modify starch by oxidation. The product, oxidized starch, has molecular weight and intrinsic viscosity less than the native starch. On the contrary, carboxyl and carbonyl contents are higher.

There are three kinds of dextrin, a product of pyroconversion of starch: British gum, white dextrin, and yellow or canary dextrin. White dextrin are made at low temperature and low pH conditions, British gum at high temperature and high pH, while yellow dextrin are made at low pH and high temperature with the addition of dilute acids such as hydrochloric or sulfuric acids. Dextrin are applied to replace gum arabic for use in the manufacture of adhesives as for sealing envelopes, gumming of papers, and for other such common adhesive applications (Wurzburg, 1987).

III. EXPERIMENTAL METHODS

3.1 Research Strategy

Both sago flour and wheat flour are starch rich materials. However, unlike the wheat flour, previous research (Sumadiwangsa, 1985) has shown that sago flour cannot be used directly as a direct replacement of wheat flour as an extender for UF-adhesives without some sort of modification or change in adhesive formulation strategy. The challenge of this research will be to modify sago flour to obtain a material that works successfully in a U-F adhesive formulation for bonding wood veneers and to determine a formulation strategy that will allow the use of sago flour in a UF adhesive formulation.

Compared to wheat flour, sago flour has larger granule size and, when mixed with water, imparts higher viscosity (Table 6). From preliminary studies, it is clear that sago flour is faster to retrograde (precipitate) and has less hydrosol compared to wheat flour.

Until now the criteria for determining the quality of an extender is still somewhat ambiguous. In this research, the property which will be explored and manipulated is viscosity, while others such as pH, water intake, and precipitate time will be determined with regard to achieving values closest to those of wheat flour.

This experiment can be categorized into 3 stages, as follows:

1. Sago modifications
2. Exploring the effect of percentage of sago (modified and unmodified) on glue viscosity and then to use the derived data to suggest optimum glue mixtures (on the basis of viscosity)

3. Comparing the bonding properties of the most suitable mixture as an extender (on the basis of viscosity) with the control wheat flour mixture.

3.1.1 Sago modification

Prior to modification, the characteristics of sago were determined and compared with a broadly accepted extender (wheat flour). To make sago usable it needs to be modified to achieve characteristics somewhat the same as those of wheat flour, especially in viscosity, pH, water intake, and precipitate time. For these purposes two methods of modifications had been carried out, first acid modification and second phosphorylation.

3.1.1.1 Acid modification

The aim of acid modification was to make the sago flour-fluid mixture less viscous than mixtures using sago in its original state. This step was designed to give the sago mixture about the same viscosity as the wheat flour mixture, and thence to make it useable by Indonesian plywood factories. The viscosity of acid modified sago depends on acid concentration used, length of reaction time, temperature of acid modification (Rohwer and Klem, 1984; and Wurzburg, 1986), and type of acid used (Wurzburg, 1986).

Higher reaction temperature, higher acid concentration, and longer time of acid modification, all resulted in lowered viscosity of starch extended products. One step of the experiment was to ascertain the optimum duration of acid modification reaction time, in order to achieve a level of fluid modified sago viscosity near that of the fluid wheat flour mixture.

3.1.1.2 Phosphorylation

The purpose of phosphorylation was to increase hydrosol level (determined by water intake), and to prolong precipitation time of sago flour (Solarek,1986). Again the water intake and precipitation time target values of each were closest to that of the wheat flour. The research will try to establish effective and appropriate conditions for sago phosphorylation. The reagents that will be employed in the research are mixtures of monosodium hydrogen phosphate (NaH_2PO_4), disodium hydrogen phosphate (Na_2HPO_4), and urea (Kerr,1960; Neukom,1958; Rutenberg & Solarek,1984). Clearly, one may not achieve exact matching of all properties; viscosity was the property given the greatest emphasis in the present study.

3.1.2 Effects of Extender and Water Contents on Viscosity and Bonding

The percentage of extender, viscosity of glue mixtures, and curing temperature contribute to the bond strength and curing rate of an adhesive in addition to adhesive formulation and catalyst. To obtain an optimum mixture of the trial extender (sago modified and unmodified) experiments have been conducted in three stages. The first stage was to seek the optimum percentage of each extender, the second stage was to seek the optimum viscosity, and the third was to explore the effect of temperature to the curing properties isothermal strength development rate and maximum strength.

Most Indonesian plywood factories spread UF adhesive mixtures at the viscosity of 1300 - 1800 cPs (Sumadiwangsa,1992), so the target of viscosity of the glue mixture in this experiment was 1500 cPs.

A higher percentage or quantity of the sample (natural, acid modified, and phosphorylated sago) needs more water to achieve the target viscosity of about 1500 cPs. In the first step of the second stage of the experiment, the purpose was to find the most suitable percentage among the five percentages of extenders investigated (at 1500 cPs in viscosity). The isothermal curing rate of them was evaluated by using an automated bonding evaluation system "ABES" (Figure 3.1) developed by Humphrey & Zavala and improved by Humphrey (1993). In brief, the temperature of the test (curing glue temperature) and the viscosity were constant, while the percentage of the sample of the mixtures were variables. There were three samples in this test: natural, acid modified, and phosphor modified sago.

After the most suitable percentage of sample had been found, if any, it was used as an arbitrary constant to find out the most suitable viscosity, instead of 1500 cPs. In this second step of the second stage, five levels of viscosity were explored to aid in choosing the most suitable viscosity of glue mixture. The tests were carried out employing the ABES to measure the rate of strength development. A Universal Testing machine was used to measure maximum bond strengths for those completely cured.

The ABES machine automatically closes to form bonds between two pieces of veneer at adjusted temperature and then after a selected curing time the machine automatically opens and the bond tested. All data related to bond strength such as pressing pressure, temperature and pressure time, overlapping area, and bond strength are automatically recorded (Humphrey, 1994).

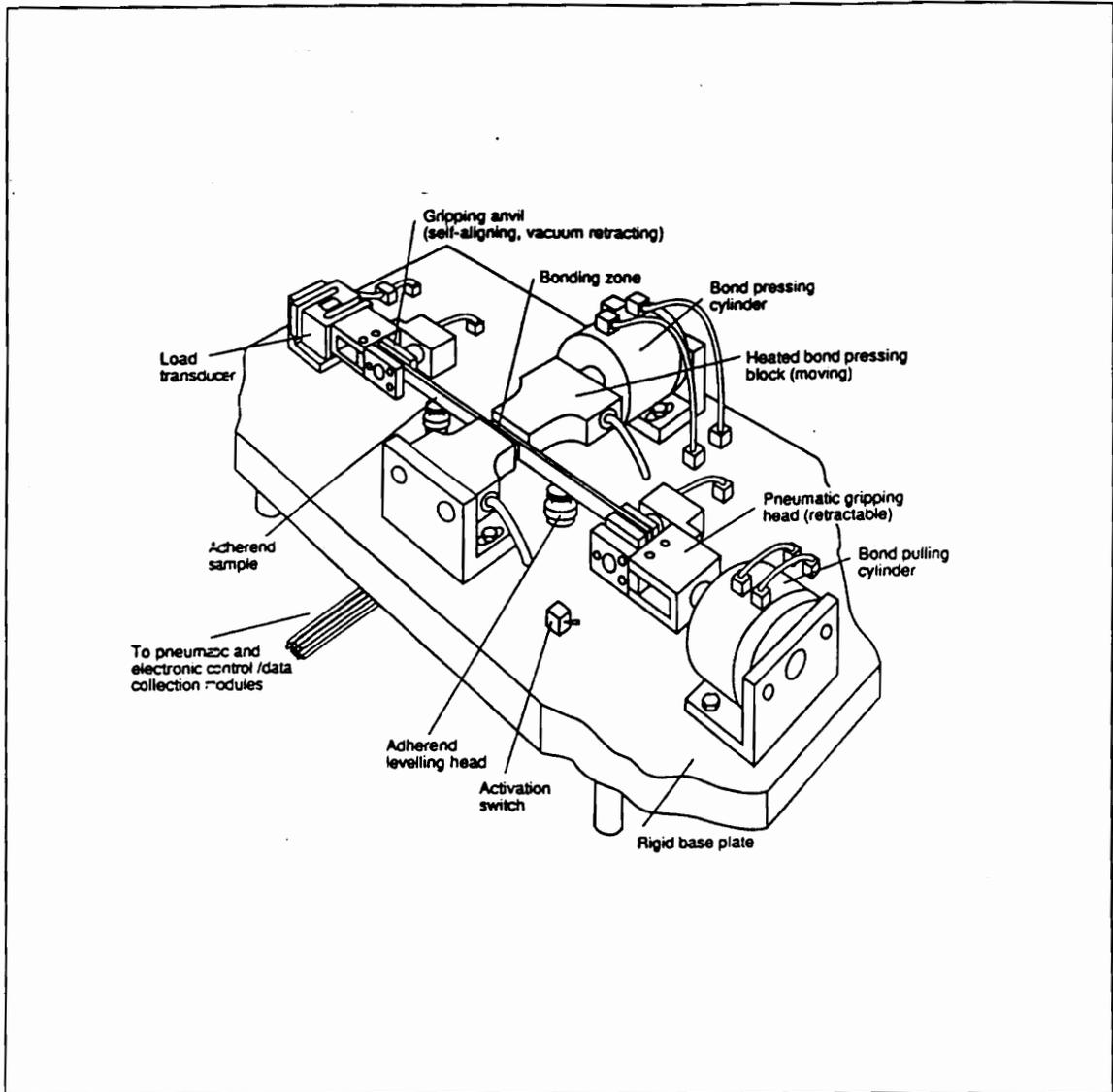


Figure 3.1 Automated Bonding Evaluation System (ABES) Machine*

* Courtesy of DR Philip E. Humprey

3.1.3 Effect of Temperature on Bonding

The values of percentage extender and viscosity that have been found in the second stage were used as arbitrary constants in this stage (third stage) of the experiment. The variable here was temperature which consists of five levels. This third stage characterized glue bonding of the tested extender. For comparison, an ordinary extender (wheat flour) were tried as well.

3.2 Experimental Design

3.2.1 Material

Sago was obtained from a small sago factory (Ata Sasmita) at Bogor, Jawa Barat, Indonesia.

Meranti veneer, 0.8 mm thickness, was obtained from a plywood factory at Banjarmasin, Kalimantan Selatan, Indonesia.

Urea-formaldehyde glue (GP 1955 UF Hardwood Plywood Resin), catalyst (GP 0210 Liquid Catalyst), and extender were received from Georgia Pacific, Resin, Inc. Albany, Oregon. UF-resin itself has 60 % resin solids.

Chemicals were ACS (American Chemical Society) reagent grade.

3.2.2 Characterization of Samples

Samples to be characterized were:

- sago unmodified
- common extender (wheat flour)
- sago modified
 - acid modified
 - phosphorylated

The procedures to characterize sago properties of this work were adopted from Smith (1967).

3.2.2.1 Moisture Content

About two grams of sample were weighed on a dish of known weight (B grams). The weight of an empty and oven dried dish was A grams. The dish was placed in an oven at 105 - 110° C. for four hours. The dish was then reweighed at one hour interval until the weight was constant (C grams). The moisture content was calculated as follows:

$$\text{Moisture Content} = \frac{B - C}{C - A} \times 100 \%$$

3.2.2.2 Bulk Density

Bulk density was determined using a 25 ml graduated cylinder. The sample was carefully poured into the cylinder (having been weighed, A gram) through a powder funnel until the top reached the 25 ml mark. The full cylinder was weighed (B gram). The bulk density (loose) was calculated as follows:

$$\text{Bulk Density, loose} = \frac{B - A}{25} \text{ grams/ml}$$

The graduated cylinder including the content was then vibrated by a vibrator until the volume was constant (D ml). The bulk density (vibrated) was calculated as follows:

$$\text{Bulk Density, vibrated} = \frac{B - A}{D} \text{ grams/ml}$$

3.2.2.3 Hot Paste Viscosity

Hot paste viscosity was determined using a Brookfield Synchro-Lectric Viscometer. About two grams of the sample was dissolved in 50 cc of water in a 80 ml glass beaker. The mixture was heated in a steam bath at 85° C. for 30 minutes while occasionally being stirred. Then the mixture was cooled. After the mixture reached ambient temperature, the viscosity was determined using an appropriate speed and spindle type, at 24° C ($\pm 1^\circ$ C).

3.2.2.4 pH

The value of pH was measured using a Corning model 106 pH meter. Before using this, the device was calibrated with a pH 7.0 buffer solution. About three grams of the sample was weighed, six ml of distilled water was added. The pH of the mixture was measured using the pH meter mentioned above.

3.2.2.5 Precipitation Time

About three grams of the sample was weighed out and mixed with six ml of distilled water in a 10 mls graduated cylinder. The top of the precipitate was measured every minute, until no more precipitation took place.

3.2.2.6 Water Intake

The water layer on a precipitate time measurement was removed by employing a pipet and weighed (C grams). The weight of the empty graduated funnel was A grams, with an added three grams of sample it weighed B grams. If the precipitate weight in grams, then the water intake was calculated as follows:

$$\text{Water intake} = \frac{C - A}{B - A} \times 100\%$$

3.3 Sago Modification

Two methods of sago modification were employed in this experiment:

1] acid modification, and 2] phosphorylation.

3.3.1 Acid Modified Sago

Sago flour was soaked in 7.5% HCl at ambient temperature. After one day, the sago flour was neutralized, washed, and dried at room temperature. After drying, the sago properties were determined. In addition to one day of soaking, there were also 3, 5, 7, and 9 days of soaking treatments.

The most suitable one of the five treatments was selected as the material for the next phosphorylation step. The seven day treatment (Lintner starch, if using potato instead of sago flour) was tested and characterized as acid modified sago from here on termed ("A-sago").

3.3.2 Phosphorylated Sago

Before phosphorylation, sago flour was treated by one day soaking in 7.5% hydrochloric acid (HCl) at room temperature. The phosphorylation was carried out using a mixture of monosodium hydrogen phosphate (NaH_2PO_4) and disodium hydrogen phosphate (Na_2HPO_4) at a ratio of one to one. The total phosphate salts added was 0.9%. Five percent of urea was also added to the mixture. (Percentages are calculated by the weight of air dry sago flour).

Na_2HPO_4 and NaH_2PO_4 each was weighed to about 0.45 grams and also weighed 5.0 grams of urea. All of these reagents were dissolved in 100

grams of distilled water. The mixture was poured into 100 grams of sago flour, mixed well and let stand for 30 minutes, while occasionally being stirred. The mixture was filtered through filter paper (VWR grade 613) using a vacuum pump, and then oven dried for 30 minutes. The mixture was phosphorylated in an oven at 140° C. for two hours. The crude phosphorylated sago, after reaching room temperature, was washed with distilled water and then air dried to obtain the phosphorylated sago from here on termed ("P-sago").

3.4 Viscosity of Adhesive Mixtures

The adhesive manufacturer proposed the formula for the glue mixture proportions; resin, catalyst, extender, and water were 1070, 75, 460, and 600 lbs, respectively. Even though, in this experiment, the amount of trial extender (natural, acid modified, and phosphorylated sago), to get certain viscosity was varied, instead of using a fixed quantity of 93.5 percent (based on resin solids).

The amounts of natural sago (N-sago) tried was 0%, 94%, 130%, and 182% by the weight of solid UF resin. UF resin itself had 60% resin solids or the resin mixture had 29.1% solid resin. The percentage of acid modified sago (A-sago) which was tried in the glue mix was 52%, 78%, and 130%. The percentage of phosphorylated sago (P-sago) was 38%, 78%, and 130% by the weight of solid UF resin. The expected viscosity was between 300 - 3000 cPs. To obtain that viscosity, water had to be added. The quantity of water for each kind and level of each percentage of sago was detailed in Table 3.1.

To 166.7% of UF resin and 11.7% of catalyst one cell of the Table 3.1 was added. For example, consider cell P12. About 13% of water (cell P12) was added to 166.7% of UF resin, it was mixed well for 30 seconds.

Then 11.7% of catalyst was added and mixed for 30 seconds. After that, 38% of natural sago (cell P12) was added, mixed well and the viscosity and pH were measured at room temperature ($24 \pm 1^\circ \text{C}$). From the collected data, the relation between water added and viscosity for each percentage of trial extender was determined. For example: using 78% of natural sago, there were five viscosity values for the level of 13%, 18%, 27%, 38%, and 52% of water. A chart for each of those relations has been drawn. The quantities of water needed to get 1500 cPs in viscosity for any of the levels of trial extender percentages was calculated from the chart.

3.5 Test of Adhesive Quality

To blend the adhesive, Georgia-Pacific Resins, Inc. provided the guidelines, as follows:

| | lb | % of mixtures | % of solid resin |
|-------------|------|---------------|------------------|
| Resin | 1070 | 48.53 | 166.67 |
| Catalyst | 75 | 3.40 | 11.68 |
| Water | 460 | 20.86 | 71.85 |
| Wheat Flour | 600 | 27.21 | 93.46 |
| Total: | 2205 | 100 | 343.46 |

Resin and water were mixed and stirred for 30 seconds. After which the catalyst was added and stirred for another 30 seconds. Finally, wheat flour (or a trial extender) was added and stirred until the mixture was homogenous.

Table 3.1 Percentage of Trial Sago and Water Added to Obtain Specific Viscosities of Adhesive Mixtures

| Water added, % | Trial sago added, % | | | | | | |
|----------------|---------------------|-----|-----|--------|-----|--------|-----|
| | 0 | 38 | 52 | 78 | 94 | 130 | 182 |
| 0 | | P11 | A21 | | N41 | | |
| 8 | | | A22 | | | | |
| 13 | | P12 | | A31 | N42 | | |
| 18 | | | A23 | A32 | N43 | | |
| 27 | | P13 | A24 | A33P31 | N44 | N51 | |
| 34 | | | | | | N52 | N61 |
| 39 | | P14 | A25 | A34P32 | N45 | A51N53 | N62 |
| 44 | | | | | | A52 | |
| 47 | | | | | N46 | N54 | N63 |
| 52 | | P15 | | A35P33 | | A53P51 | |
| 60 | | | | | | N55 | N64 |
| 65 | | | | P34 | | A54P52 | N65 |
| 78 | | | | P35 | | A55P53 | N66 |
| 92 | | | | | | P54 | |
| 104 | | | | | | P55 | |

Note : Percentage is calculated by the weight of UF solid resin.

N: natural sago

A: acid modified sago

P: phosphorylated sago

In the experiments, the amount of resin, catalyst, water and wheat flour (or a trial extender) were reduced to 6.4, 0.4, 2.3 and 3.0 grams, respectively.

Wheat flour was replaced by trial extenders (natural, acid modified and phosphorylated sago). Each amount was adjusted according to the percentage necessary.

To achieve the target viscosity level, an appropriate amount of water was added to the fixed amount of trial extender, resin, and catalyst.

All calculated percentages of materials were based on solid resin weight.

The maximum bond strength (400 seconds at 95° C.) and the rate of curing adhesive of each adhesive mixture was measured. To measure the rates of curing the ABES machine was used. To measure maximum bonding strengths a Universal Testing machine was employed.

The veneer strip samples (veneer specimens) were thin enough (0.8 mm) to enable the glue-line to rapidly reach the target temperature (about 20 seconds) once placed in the ABES system. In this experiment, the size of adherents were 97 mms in length, and 20 mms in width. The glue area was 20 mm by 3 mm (Humprey, 1994).

In this experiment, bond forming pressure was keep constant at 5.7 MPa. To investigate the effect of adhesive component ratios, and viscosities of fluid adhesive mixtures, the temperature was keep constant at 95° C. Pressing time range between 20 to 180 seconds, since up to 20 seconds was needed to achieve the target glue-line temperature within $\pm 1^{\circ}\text{C}$.

There were two steps in the ABES testing program. First to set for a group of samples of each trial extender treatments was tested, and secondly every sample was tested. To form each bond the machine was set to the desired pressing temperature and pressure, and the length of the overlapping area to glue. A group file name was entered into the computer together with the length of curing time (in seconds) desired. To prepare each sample two pieces of veneer where one end of a veneer was spread with a glue sample. The sample was inserted into the compressing compartment on the ABES machine, the machine started, the sample named and the bond strength data collected.

3.5.1 Effect of Ratio of Trial Extenders on Adhesive Bonding

The viscosity of adhesive mixtures for each experiment was made constant at 1500 cPs. Such a viscosity was used by most plywood factories in Indonesia. The temperature used to cure the test specimen was 95° C. Formulation of the test adhesive mixtures at this stage of the research was done as follows. To urea-formaldehyde (GP-1955) resin (6.4 grams liquid resin contains 3.84 grams of resin solids) was added the specified amounts of water and then mixed well for about 30 seconds. To this mixture was added 0.4 grams of catalyst (GP-0210), which was again mixed well for about 30 seconds. Then to the resulting mixture was added a specified amount of each trial extender (N-sago, A-sago, or P-sago), proportionate to the water amounts below. The resultant final viscosity of each adhesive mixture was 1500 cPs. The mixture was mixed well until homogenous. The adhesive mixture was then ready to spread on a veneer surface specimen to determine the adhesive quality: rate of bond strength and maximum bond strength. The percentage of natural sago employed in each respective experiment were 50, 83, 108, 133.3 and 150%,

with 3.7, 14.0, 21.7, 28 and 33.8% water added, respectively. All percentages were based on resin solids. The percentage of acid modified sago and water were 50, 67, 83, 108, 125, and 4.2, 11.0, 18.0, 28.2, 36.7%, respectively, also based on resin solids. The percentage of phosphorylated sago and water were 34, 58, 83, 108, 133 and 7.5, 20.3, 33.8, 38.0 and 54.5% based on resin solids, respectively.

3.5.2 Effect of Viscosity on Adhesive Bond Quality

Since there was no obvious optimum percentage for each extender which was tested in the previous experiments, the percentage was chosen in light of the adhesive manufacturers' guidelines. The extender amount was 93.46% based on resin solids. The viscosities ranged from about 10 up to near 10000 cPs. The composition of the glue mixtures for natural sago (N-sago) range from 0 to 200% to make viscosities range from 8650 to 15.5 cPs (Table 3.2). Acid modified sago (A-sago) as a trial extender needed water from 10 to 250% with the viscosities ranging from 9300 to 13 cPs (Table 3.3). Phosphorylated sago (P-sago) needed water from 18 to 400% to make the viscosities from 9400 to 11.5 cPs as shown in Table 3.4. Each composition was used in an attempt to two parallel grain oriented to glue meranti veneers, depending on the curing time. The curing temperature was maintained constant at 95° C. The rate of curing (ROC) and the maximum bond strength (MBS) data were collected by using the ABES and Universal Testing Machine respectively.

From these tests it was determined which adhesive viscosity gave the respective test specimens the best rate of curing and maximum bond strength for each of the kinds of sago flour extender.

3.5.3 Effect of Curing Temperature

According to the results of the above experiments, the adhesive formulation used for curing temperature tests for each kind of the extenders, are shown below:

| Materials | natural | <u>Sago modified</u> | | Wheat flour |
|-----------------|---------|----------------------|----------|-------------|
| | | acid | phosphor | |
| UF resin, grams | 6.08 | 6.08 | 6.08 | 6.08 |
| Catalyst, grams | 0.43 | 0.43 | 0.43 | 0.43 |
| Extender, grams | 3.38 | 3.38 | 3.38 | 3.38 |
| Water, grams | n | a | p | wf |

Note: n, a, p, and wf will be employed from the last experiment to achieve the various viscosities shown in Table 3.2, 3.3, and 3.4.

The curing rate and the maximum bonding strength of each formulation (sample codes N21V - P35V) were tested employing the ABES and the Universal Testing machine. The temperature employed were 85, 95, 105, 115 and 125° C.

Table 3.2 Formulation of the Test UF Adhesive Mixtures with Natural Sago as an Extender

| Sample Code | N21V | N29V | N30V | N31V | N37V |
|----------------------|--------|--------|--------|--------|-------|
| UF resin, grams | 6.08 | 6.08 | 6.08 | 6.08 | 6.08 |
| Catalyst, grams | 0.43 | 0.43 | 0.43 | 0.43 | 0.43 |
| Natural sago, grams | 3.38 | 3.38 | 3.38 | 3.38 | 3.38 |
| Water, grams | 7.29 | 2.62 | 2.18 | 1.83 | 0 |
| Total weight, grams | 17.18 | 12.51 | 12.07 | 11.72 | 9.98 |
| Water, added % 1) | 199.84 | 71.82 | 59.76 | 50.16 | 0.0 |
| 2) | 42.43 | 20.94 | 18.06 | 15.61 | 0.0 |
| Water contents, % 1) | 266.51 | 138.49 | 126.43 | 116.83 | 66.67 |
| 2) | 56.59 | 40.37 | 38.19 | 36.35 | 24.35 |
| Extender, % 1) | 92.65 | 92.65 | 92.65 | 92.65 | 92.65 |
| 2) | 19.67 | 27.02 | 28.00 | 28.84 | 33.87 |
| Solid resin, % 2) | 21.23 | 29.16 | 30.22 | 31.13 | 36.55 |
| Viscosity, cPs | 15.5 | 132 | 640 | 1350 | 8650 |

1) based on solid resin

2) based on total mixtures

Table 3.3 Formulation of the Test UF Adhesive Mixtures with Acid Modified Sago as an Extender

| Sample Code | A19V | A26V | A30V | A33V | A36V |
|----------------------|--------|--------|--------|-------|-------|
| UF resin, grams | 6.08 | 6.08 | 6.08 | 6.08 | 6.08 |
| Catalyst, grams | 0.43 | 0.43 | 0.43 | 0.43 | 0.43 |
| A-sago, grams | 3.38 | 3.38 | 3.38 | 3.38 | 3.38 |
| Water, grams | 9.11 | 4.13 | 2.31 | 1.21 | 0.38 |
| Total | 19.0 | 14.02 | 12.2 | 11.1 | 10.27 |
| Water, added % 1) | 250.07 | 113.37 | 63.41 | 33.21 | 10.43 |
| 2) | 47.95 | 29.23 | 18.93 | 10.90 | 3.70 |
| Water contents, % 1) | 316.39 | 170.04 | 130.08 | 99.88 | 77.10 |
| 2) | 60.75 | 46.80 | 38.87 | 32.81 | 27.38 |
| Extender, % 1) | 92.65 | 92.65 | 92.65 | 92.65 | 92.65 |
| 2) | 17.79 | 24.11 | 27.70 | 30.45 | 32.91 |
| Solid resin, % 2) | 19.20 | 26.02 | 29.90 | 32.86 | 35.52 |
| Viscosity, cPs | 13 | 60 | 300 | 1800 | 9300 |

1) based on solid resin

2) based on total mixtures

Table 3.4 Formulation of the Test UF Adhesive Mixtures with Phosphorylated Sago as an Extender

| Sample Code | P15V | P27V | P30V | P33V | P35V |
|---------------------|--------|--------|--------|-------|-------|
| UF resin, gram | 6.08 | 6.08 | 6.08 | 6.08 | 6.08 |
| Catalyst, gram | 0.43 | 0.43 | 0.43 | 0.43 | 0.43 |
| P-sago, gram | 3.38 | 3.38 | 3.38 | 3.38 | 3.38 |
| Water | 14.57 | 3.64 | 2.19 | 1.09 | 0.66 |
| Total | 24.46 | 13.53 | 12.08 | 10.98 | 10.55 |
| Water, added % 1) | 399.40 | 99.78 | 60.03 | 29.88 | 18.09 |
| 2) | 59.57 | 26.90 | 18.13 | 9.93 | 6.26 |
| Water content, % 1) | 466.06 | 166.45 | 126.70 | 96.55 | 84.76 |
| 2) | 69.51 | 44.88 | 36.26 | 32.08 | 29.31 |
| Extender, % 1) | 92.65 | 92.65 | 92.65 | 92.65 | 92.65 |
| 2) | 13.82 | 24.98 | 27.98 | 30.78 | 32.04 |
| Solid resin, % 2) | 14.91 | 26.96 | 30.20 | 33.22 | 34.58 |
| Viscosity | 11.5 | 120 | 560 | 3100 | 9400 |

1) based on solid resin

2) based on total mixtures

IV. RESULTS AND DISCUSSIONS

4.1 Properties of Natural Sago and Wheat Flour Extender

Almost all of the properties of the sago starch and wheat flour extender that have been tested differ, especially viscosity, water intake, and retrogradation (on time) (Table 4.1). These three are important properties to examine in view of the role each plays in adhesive performance.

Control of viscosity is the most important function of an extender. Klein (1980) has said that an extender should be able to control viscosity as well as to reduce UF-resin consumption. In practical terms, that means the viscosity of an extender needs to be higher than UF resin itself or than the final glue mixture. The hot paste viscosity of four percent natural sago (6533 cPs) is greatly higher than the wheat flour (8.6 cPs) (Table 4.1). Knight (1969) has previously noted that sago flour is larger in molecular size, and that it has a higher granular size and viscosity compared to other starchy materials.

Water intake is the second important property to examine in the role of extender substitutions. An extender has to be a hydrophilic material, since UF-resins are water borne adhesives. The water intake of natural sago is 160.3 percent, while that of wheat flour is 231.2 percent. The water intake difference is due to differences in the molecular size of sago compared to wheat flour. In modifying sago flour to have a higher water intake, one method could

be to reduce its molecular size. One method to do this is by acid catalyzed cleavage of the inter-glucosyl acetal linkages. Such an acid treatment would have to be carefully controlled to achieve the desired reduction in molecular size and not complete hydrolysis to monomer units (glucose).

Table 4.1 Comparison Between Sago Starch and Wheat Flour Properties*

| Properties | Natural sago | Wheat flour |
|--|--------------|-------------|
| Moisture contents, % | 14.65 | 12.06 |
| pH | 4.58 | 5.97 |
| Retrogradation, - minutes | 31.3 | 232.3 |
| - % volume | 52.1 | 79.3 |
| Weight/volume - loose, g.cm ⁻³ | 0.675 | 0.429 |
| - dense, g.cm ⁻³ | 0.856 | 0.583 |
| Viscosity, cPS | 6533 | 8.6 |
| Water intake, g/g, % | 160.3 | 231.2 |

* Average of 3 replications

Retrogradation or precipitation time is another measure of an extenders' property. It is essential that an extender remains suspended within the adhesive. If an extender settles (retrogrades) too fast, the glue mix will become inhomogeneous and excess water in spots will cause problems with adhesion. In a practical sense, such spots of water in plywood adhesion are a leading cause of panel blows. Thus, localized spots of excess moisture need to be avoided. In

addition, at all costs, such localized excess water will also cause over penetration of the adhesive and reduce the adhesive cure rate and thus leading to what is (Karchesy,1994) referred to as " glue line wash out" in the plywood industry. This situation also needs to be avoided at all costs. Natural sago has a faster retrogradation time (31.3 minutes) compared to wheat flour (232.2 minutes) (Table 4.1)

Several methods are available for the modification of sago to give it properties similar to wheat flour with regard to viscosity, retrogradation, and water intake. Specifically, viscosity needs to be decreased, retrogradation stopped, and water intake increased. If these goals can be achieved, then there is a chance to achieve a useable adhesive formulation for urea-formaldehyde with a sago flour extender. Probably the two best methods of modifications theoretically suitable for these purposes are acid modification and phosphorylation. Both methods have been explored to determine the suitability of modifying sago flour for use as an extender for UF adhesives with wood veneers.

4.2 Acid Modification of Sago

The aim of acid modification is to make the sago flour a fluid mixture which is less viscose than the mixture using sago in its original state. Viscosity of acid modified sago depends on the acid concentration used to modify the starch, reaction time, temperature of the modification reaction

(Rohwer and Klem, 1984, and Wurzburg, 1986), as well as the type of acid used (Wurzburg, 1986). In this experiment 7.5 % HCl solution is used as a modification reagent. It has been shown that acid modified starch does not change the physical form of starch, insolubility in cold water, and birefringence properties according to Shildneck and Smith, 1967). However, less drastic but subtle changes do occur. Compared to the parent starch, acid modified starch has a) lower hot paste viscosity, b) a higher alkali number, c) higher ratio of cold to hot paste viscosity, d) higher osmotic pressure, e) higher gelatinization temperature, f) higher critical absorption of sodium hydroxide, g) lower intrinsic viscosity, h) lower iodine affinity, and i) less granule swelling during gelatinization (Shildneck and Smith, 1967).

In acid modification, the acid mainly attacks the amorphous region of the starch granule since that portion is more accessible and this leads to a reduction in viscosity of the starch paste (Rowner and Klem, 1984).

Acid modification experiments done on sago flour indicate that viscosity and water intake were successfully altered (Table 4.2). Both properties near those of wheat flour were obtained. After one day of soaking in 7.5% HCl, sago has the viscosity (6.9 cPs) which is near that of wheat flour (8.6 cPs). After more than one day of soaking the viscosity persistently decreases, and after nine days of soaking the viscosity is a very low 2.1 cPs (Table 4.2).

The longer the acid soaking, the greater the water intake in acid modified sago. After seven and nine day periods of soaking the water intake of acid modified sago (203.5 and 202.8 g/g) is near the same level of wheat flour at 231.2 g/g (Table 4.2).

After seven and nine day periods of soaking, the retrogradation and the other properties seemed to remain constant as they did with natural sago (Table 4.2).

4.3 Phosphorylated Sago

Phosphorylation of natural sago increased its pH, retrogradation, water intake, and viscosity (>10000 cPs) as can be seen in Table 4.3. These results are consistent and similar to those reported by Kerr (1960). Since the viscosity of phosphorylated sago (> 10,000 cPs) was unusable in this range, it was decided to explore phosphorylation effects on one day acid treated sago. From Table 4.3 one can see that this combination of treatments gave material with an acceptable viscosity value of 13.0 cPs. In fact, all of the values in Table 4.3 for the phosphorylated-acid soaked sago, compare very favorably to those of the wheat flour standard also shown in Table 4.3. The retrogradation value of 122 minutes is about half the time length time for wheat flour (232 minutes), but this length of time is judged to be an adequate time for any adhesive mixture assembly time.

Table 4.2 Sago Starch Properties After Several Days Soaking in 7.5 % HCl Acid*

| Properties | HCl soaking period (days) | | | | | Wheat Flour |
|----------------|---------------------------|-------|-------|-------|-------|----------------|
| | 0 | 1 | 3 | 7 | 9 | |
| M. C. (%) | 14.6 | 14.3 | 13.7 | 14.3 | 13.3 | 12.1 |
| pH | 4.58 | 4.93 | 4.93 | 4.89 | 5.01 | 5.97 |
| Retrogradation | | | | | | |
| - minutes, | 31.3 | 36.3 | 36.7 | 38.3 | 39.0 | 232.3 |
| - % volume | 52.1 | 53.0 | 57.2 | 59.0 | 59.4 | 79.3 |
| Weight/volume | | | | | | |
| - loose | .675 | .711 | .695 | .692 | .677 | .429 |
| - dense | .856 | .861 | .826 | .865 | .848 | .583 |
| Viscosity, cPs | 6533 | 6.9 | 3.1 | 2.3 | 2.1 | 8.6 |
| Water intake,% | 160.3 | 162.7 | 182.1 | 203.5 | 202.8 | 231.2 |

* Average of 3 replications

4.4 Viscosity of Glue Mixtures

The viscosity and pH of glue mixtures using various extender mixtures are detailed in Appendix 4.4 - 4.6. The influence of varying amounts of water on viscosity with selected trial extenders is shown at Figure 4.1 for natural sago (N-sago), Figure 4.2 acid modified sago (A-sago), and Figure 4.3 phosphorylated sago (P-sago). As one would

Table 4.3 Phosphorylated Sago Properties from Natural and One Day Acid Soaking*

| Properties | Sago nat. | <u>Phosphorylated sago</u> | | Wheat flour |
|-----------------|-----------|----------------------------|------------|-------------|
| | | natural | acid soaks | |
| M.C., % | 14.65 | 13.35 | 13.15 | 12.06 |
| pH | 4.58 | 6.87 | 5.85 | 5.97 |
| Retrogradation, | | | | |
| - minutes | 31.3 | 71.3 | 121.7 | 232.3 |
| - volume, % | 52.1 | 63.4 | 74.2 | 79.3 |
| Weight/volume | | | | |
| - loose | .675 | .610 | .660 | .429 |
| - dense | .856 | .811 | .772 | .583 |
| Viscosity, cPs | 6533 | >10000 | 13.0 | 8.6 |
| Water intake, % | 160.3 | 192.3 | 224.7 | 231.2 |

* Average of 3 replications

expect, increasing water content decreases viscosity as exhibited by these figures. The regressions are exponential with the equations as follows:

| <u>Sago samples</u> | <u>% solid resin weight</u> | <u>Equations</u> |
|---------------------|-----------------------------|---------------------------------------|
| Natural sago-A | 94 | $Y = \text{Exp} (-0.028 X) * 1808.3$ |
| natural sago-B | 130 | $Y = \text{Exp} (-0.054 X) * 6939.8$ |
| natural sago-C | 182 | $Y = \text{Exp} (-0.051 X) * 14588.9$ |
| acid modified | 52 | $Y = \text{Exp} (-0.062 X) * 2299.6$ |
| acid modified | 78 | $Y = \text{Exp} (-0.046 X) * 4156.5$ |
| acid modified | 130 | $Y = \text{Exp} (-0.039 X) * 7764.7$ |
| phosphorylated | 38 | $Y = \text{Exp} (-0.061 X) * 2733.0$ |
| phosphorylated | 78 | $Y = \text{Exp} (-0.045 X) * 5693.2$ |
| phosphorylated | 130 | $Y = \text{Exp} (-0.040 X) * 17574.3$ |
| Without | 0 | $Y = \text{Exp} (-0.036 X) * 377.7$ |

From Figures 4.1, 4.2, and 4.3 one can see the similar curves for viscosity vs. amount of water added to each trial extender mix. Figure 4.1 shows the curves for the natural sago/adhesive mixtures N-sago-A, N-sago-B and N-sago-C.

Given the same amount of added water, N-sago-A has the lowest viscosity while N-sago-C has the highest. N-sago-B is intermediate. For example, from Figure 4.1 one can see that at 50% added water to the adhesive mix, N-sago-C gave a viscosity of 230 cPs while N-sago-B gave 470 cPs and N-sago-A had a viscosity of 900 cPs.

These results are readily understandable when one considers the increasing proportion of sago extender in each adhesive mixture. In its original formulation N-sago-A mixture has a 0.94 to 1.00 ratio of starch to adhesive, N-sago-B 1.3 to 1.00, and N-sago-C 1.82 to 1.00. Thus with an increasing amount of N-sago in the trial mixture, at the same amount of water, the adhesive mixture will have a higher viscosity.

To re-emphasize, the relationship of viscosity to the amount of water and sago extender follows the curves shown in Figure 4.1 to 4.3. These results are in agreement with similar results obtained by Neukon (1958).

In Figure 4.2, A-sago-A has the ratio 0.52 to 1.00 of acid modified sago to adhesive, A-sago-B 0.78 to 1.00 and A-sago-C 1.30 to 1.00. In Figure 4.3 the ratio of phosphate modified sago to adhesive mix is 0.38 to 1.00 for P-sago-A, 0.78 to 1.00 for P-sago-B and 1.30 to 1.00 for P-sago-C.

Figure 4.4 compares the relative change in viscosity with increasing percentages of water for natural, acid treated and phosphorylated sago extenders, where each extender type is in the same proportion in the adhesive mixture (1.3 to 1.0 extender to U-F adhesive). These are designated N-sago-B, A-sago-C, and P-sago-C. Since each sago flour type is in the same proportion, one can compare the effects of modification on water percent vs. viscosity change.

In the same fashion as N-sago, A-sago and P-sago also increase in viscosity when the amount of trial extender are increased. However, the increase of viscosity of A-sago is higher than that of N-sago, and the P-sago is even higher as shown in Figure 4.4.

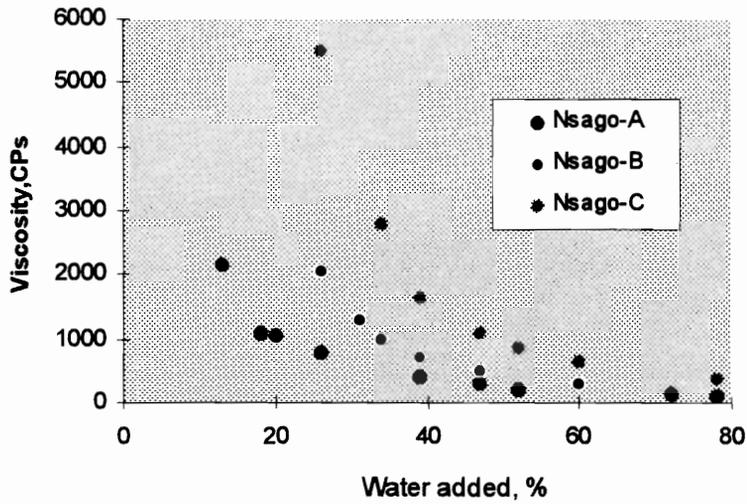


Figure 4.1. The effect of proportions of natural sago and water on viscosity of adhesive mixtures.

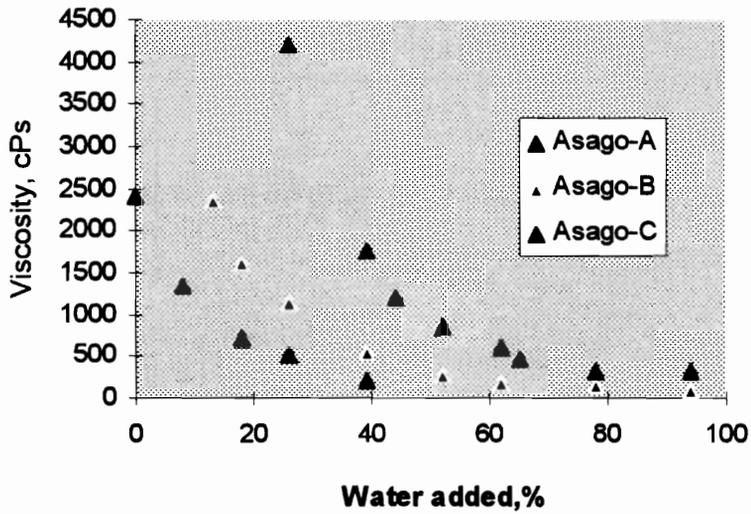


Figure 4.2. The effect of proportions of acid-modified sago and water on viscosity of adhesive mixtures.

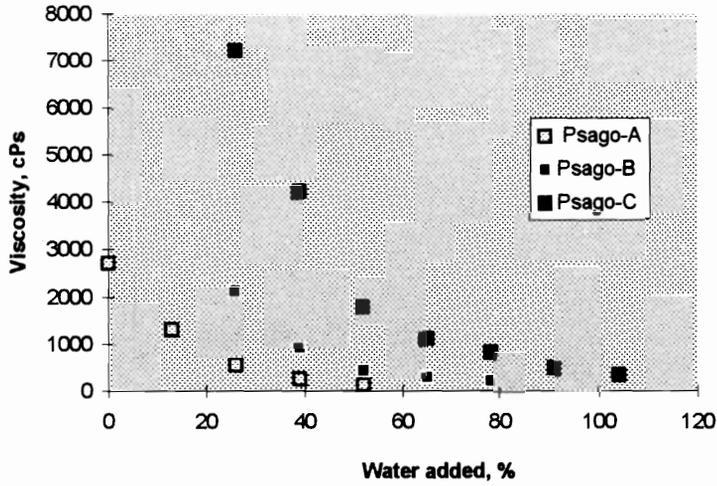


Figure 4.3. The effect of proportions of phoshorylated sago and water on viscosity of adhesive mixtures.

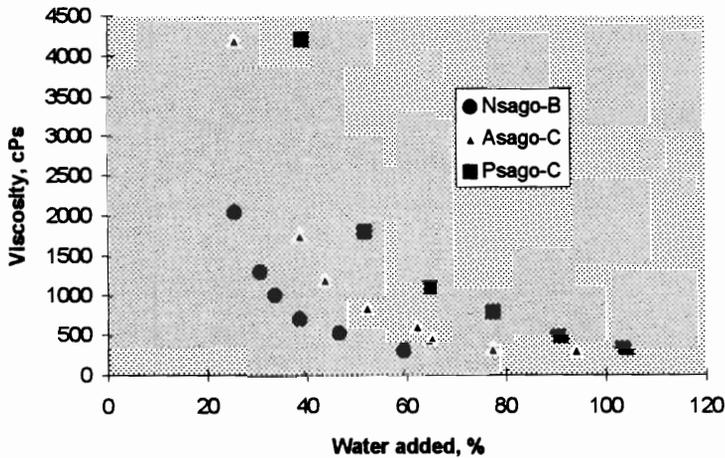


Figure 4.4. The effect of proportions of trial sago and water on viscosity of adhesive mixtures.

In order to reach 1000 cPs viscosity, 94% N-sago needs a total amount of water of about 21%; 130% N-sago needs 36%, and 182% N-sago needs 52% water (Figure 4.1). To obtain the same degree of viscosity, increasing percentage of N-sago needs more added water for each formulation (Figure 4.1). A-sago and also P-sago have the same characteristic as N-sago shown above. A-sago, however, needs more water than N-sago, and P-sago even more water than A-sago to get the same viscosity at the same proportions of materials (Figure 4.4). These results suggest that for the purpose of extending a urea-formaldehyde resin, A-sago is better than N-sago, but P-sago is the best.

To obtain a viscosity of more than 1000 cPs, the effect of water content is more sensitive. A small quantity of water added greatly increases the viscosity of all the trial extenders (Figure 4.4).

Decreasing water of N-sago by about 10% (from 35 to 25%) increased the viscosity from 1000 cPs to 2050 cPs. A-sago and P-sago at least has the same characteristic on decreasing water as N-sago (Figure 4.4). All of the three specific sago modifications in Figure 4.4 are suitable for the next step in adhesive formulation in this research since all can be formulated with a viscosity of 1500 cPs. Figure 4.5 summarizes the relationship between extender content (% of urea-formaldehyde resin) vs. added water for the three types of sago flour.

At 100% N-sago in a glue mixture, to reach a viscosity of 1500 cPs need a total water about 19%, A-sago needs 28%, and P-sago needs 41% water (Figure 4.5). At 130% N-sago, A-sago, P-sago to reach 1500 cPs on viscosity needs 28, 42, and 67% water, respectively. To get the same viscosity at the same amount of trial extender, A-sago needs water about 1.5 times of N-sago, and P-sago needs twice as much water as N-sago. Again for the purpose of extension A-sago is better than N-sago, and P-sago is the best. The amount of water needed in the adhesive mixture blended with P-sago is very similar to that required by industrial weat flour which is now used in Indonesia (Sumadiwangsa, 1985).

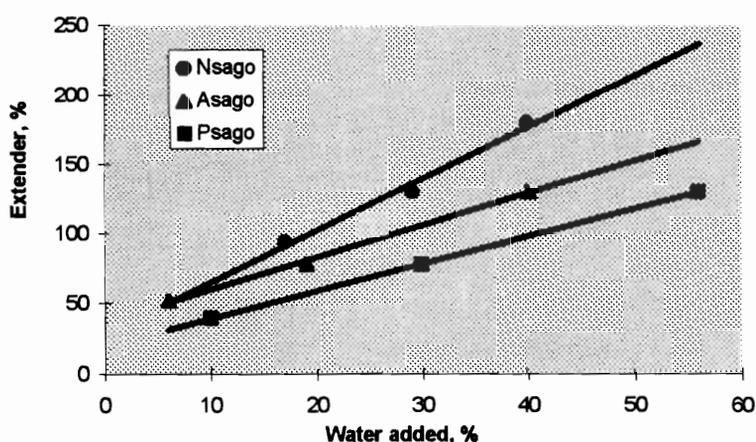


Figure 4.5. The correlation between trial extenders and proportion of water in adhesive mixtures to get 1500 cPs viscosity.

4.5 The Effect of Extender Ratio on Glue Bonding

For an adhesive mixture to be readily useful in the present industrial situation in Indonesia, it should have a viscosity between 1300 cPs and 1800 cPs. For this reason a target viscosity of 1500 cPs was chosen to develop adhesive mixtures using the sago flours. Figure 4.5 shows the latitude of water that can be added to N-sago, A-sago, and P-sago while maintaining a target viscosity of 1500 cPs. In these cases the percent of extender (compared to the adhesive polymer on a weight basis) ranged from about 25 to 230 percent depending on the exact extender used. The added water ranged from about 8 to over 50%. So, one can see quite a bit of latitude is available by varying either the amount of extender and/or water to achieve a final adhesive mixture with 1500 cPs.

Figure 4.6 compares the experimental rate of adhesive mixture curing for N-sago, A-sago, and P-sago when blended into an adhesive mixture of 1500 cPs at varying extender concentrations which range from about 30 to 150 percent by weight of the adhesive polymer. Some experimental error is noticeable in the N-sago formulations. However, in general in all cases it seems that the curing rate is not significantly altered by changing the extender ratio. It might be speculated that P-sago shows a slightly lower curing rate than the other two sago flours because the phosphate groups

may compete for reaction with the adhesive polymer methylol groups which are needed in polymerization. However this prediction needs to be further researched.

In theory the rate of adhesive cure for a urea-formaldehyde resin should be independent of the extender amount within limits and depend on factors such as adhesive polymer molecular weight, number of methylol groups, amount of water present, amount of catalyst, and temperature.

Table 4.4 summarizes the variables for each adhesive mixture used to gather the experimental data shown in Figure 4.6.

It is obvious that there are no maximum peaks, neither of curing rate, nor of maximum bond strength for all of the trial extenders added into the glue mixtures (Table 4.5 and Figures 4.6 and 4.7).

Table 4.5 briefly shows the equations and R-squares of linear regressions of curing time (in seconds) on the bond strength (in MPa) and maximum bond strength (in MPa) of natural, acid modified and phosphorylated sago for each adhesive mixture formulated in Table 4.4.

Figure 4.7 compares maximum bond strength of N-sago, A-sago, and P-sago when varied in the adhesive formulations from about 50 to 150% of the urea-formaldehyde resin polymer. The amount of U-F resin was constant. Within, experimental error, again, there is no significant change or

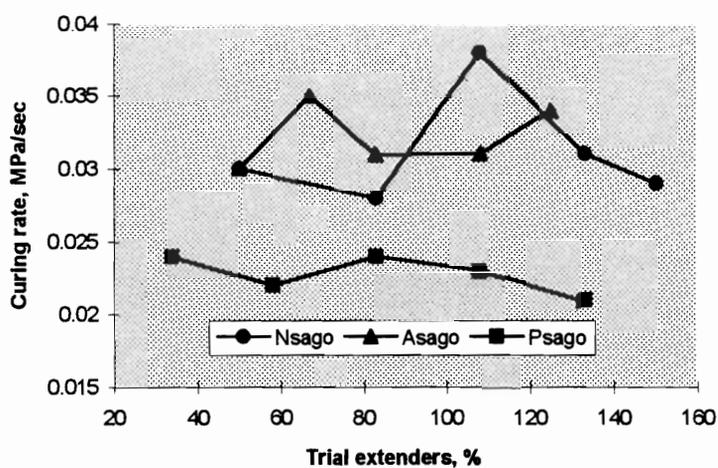


Figure 4.6. Curing rate of adhesive mixtures blended with several percentages of trial extenders.

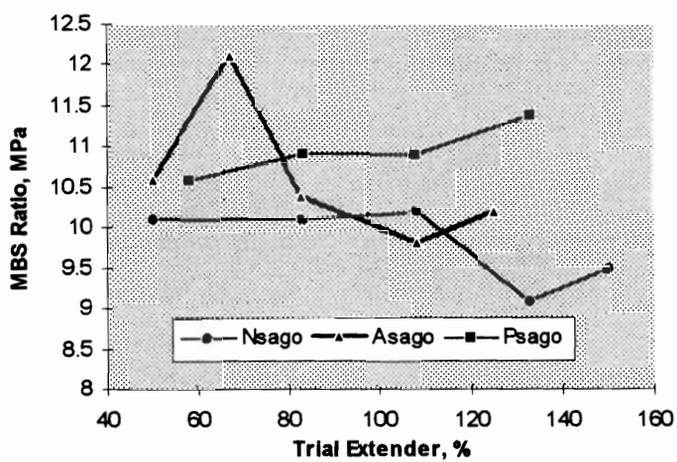


Figure 4.7. Maximum bond strength of adhesive mixtures blended with several percentages of trial extenders.

a peak in maximum bond strength. This suggests that within these parameters, the maximum bond strength is determined by the U-F resin and not by changes in the extender amounts. This is again in the agreement with the concept that the extender is not involved in adhesive polymer curing directly, but is merely acting in other capacities, eg. preventing dry out of adhesive mixture, controlling viscosity and acting as a filler (Karchesy, 1995).

Table 4.5 summarizes the regression data and maximum bond strengths for natural, acid modified and phosphorylated sago extenders. As can be seen in the Table 4.5 maximum bond strength of adhesive formulations using the natural sago ranged from 9.1 to 10.2 MPa. Those for acid modified sago extended adhesives ranged from 9.8 to 12.1 MPa and the phosphorylated sago, 10.6 to 12.1 MPa. The modified sago flour extended adhesives showed slightly higher maximum bond strengths under the conditions employed here.

Regression analysis of correlation between percentage of trial extender and maximum bond strength are shown in Table 4.6.

Table 4.4 Compositions of Urea-formaldehyde Adhesives
Mixed With Trial Extenders at 5 Ratio Levels

| | Sample number | | | | |
|-----------------------|---------------|-------|-------|-------|-------|
| | 1 | 2 | 3 | 4 | 5 |
| Viscosity, cPs | 1500 | 1500 | 1500 | 1500 | 1500 |
| UF resin, grams | 6.1 | 6.1 | 6.1 | 6.1 | 6.1 |
| Catalyst, grams | 0.4 | 0.4 | 0.4 | 0.4 | 0.4 |
| Natural Sago | | | | | |
| Nsago, % of | 50 | 83 | 108 | 133 | 150 |
| Nsago, grams | 1.82 | 3.03 | 3.94 | 4.85 | 5.47 |
| Added water, % of | 3.7 | 14.0 | 21.7 | 28.0 | 33.8 |
| Added water, grams | 0.13 | 0.51 | 0.79 | 1.02 | 1.23 |
| Total weight, grams | 8.45 | 10.14 | 11.23 | 12.37 | 13.20 |
| Solid resin, % | 43.2 | 36.0 | 32.5 | 29.5 | 27.6 |
| Acid Modified | | | | | |
| Asago, % of | 50 | 67 | 83 | 108 | 125 |
| Asago, grams | 1.82 | 2.44 | 3.03 | 3.94 | 4.56 |
| Added water, % of | 4.2 | 11.0 | 18.0 | 28.2 | 36.7 |
| Added water, grams | 0.15 | 0.40 | 0.66 | 1.03 | 1.34 |
| Total weight, grams | 8.47 | 9.34 | 10.49 | 11.47 | 12.4 |
| Solid resin, % of | 43.07 | 39.06 | 34.77 | 31.80 | 29.4 |
| Phosphorylated | | | | | |
| Psago, % of | 34 | 58 | 83 | 108 | 133 |
| Psago, grams | 1.24 | 1.83 | 3.03 | 3.94 | 4.85 |
| Added water, % of | 7.5 | 20.3 | 23.8 | 28.0 | 34.5 |
| Added water, grams | 0.27 | 0.74 | 0.87 | 1.02 | 1.26 |
| Total weight, grams | 8.01 | 9.07 | 10.40 | 11.46 | 12.61 |
| Solid resin, % of | 45.54 | 40.22 | 35.08 | 31.83 | 28.93 |

Table 4.5 The Influence of Extender Ratio on Rate of Curing and Maximum Bonding Strengths

| Trial Extender Sample # | Regressions Equation | R-squares | Max. Bond Strength, MPa* |
|----------------------------|------------------------|-----------|--------------------------|
| Natural sago | | | |
| 1 | $Y = -0.548 + 0.030 X$ | 92.3 | 10.1 |
| 2 | $Y = -0.608 + 0.028 X$ | 93.2 | 10.1 |
| 3 | $Y = -1.051 + 0.038 X$ | 97.4 | 10.2 |
| 4 | $Y = -0.516 + 0.031 X$ | 92.2 | 9.1 |
| 5 | $Y = -0.485 + 0.029 X$ | 95.5 | 9.5 |
| Acid Modified Sago | | | |
| 1 | $Y = -0.736 + 0.030 X$ | 93.7 | 10.6 |
| 2 | $Y = -0.418 + 0.035 X$ | 94.7 | 12.1 |
| 3 | $Y = -0.646 + 0.031 X$ | 94.3 | 10.4 |
| 4 | $Y = -0.323 + 0.031 X$ | 97.3 | 9.8 |
| 5 | $Y = -0.814 + 0.034 X$ | 91.9 | 10.2 |
| Phosphorylated Sago | | | |
| 1 | $Y = -0.448 + 0.024 X$ | 92.2 | 10.6 |
| 2 | $Y = -0.347 + 0.022 X$ | 97.4 | 10.9 |
| 3 | $Y = -0.111 + 0.024 X$ | 92.8 | 10.9 |
| 4 | $Y = -0.167 + 0.023 X$ | 95.1 | 11.4 |
| 5 | $Y = -0.053 + 0.021 X$ | 89.9 | 12.1 |

* Average of 5 replications
 Y = Bonding strength, MPa
 X = Curing time, seconds

Table 4.6 Regression between Percentage of Trial Extenders on Rate of Curing and Maximum Bond Strength

| | Intercept | Slope | Correlation coefficient |
|-----------------------|-----------|--------|-------------------------|
| Natural- | | | |
| Curing rate | 30.335 | 8.251 | 0.083 |
| MBS | 10.711 | -8.690 | -0.721 |
| Acid modified | | | |
| Curing rate | 30.468 | 0.020 | 0.575 |
| MBS | 12.063 | -0.017 | -0.575 |
| Phosphorylated | | | |
| Curing rate | 24.165 | -0.020 | -0.793 |
| MBS | 10.004 | 0.014 | 0.941 |

Natural sago and acid modified sago both have two secondary hydroxyl groups and one primary hydroxyl group on the extending glucosyl moieties (i.e., C-2, C-3, and C-6). Such hydroxyl groups are important in hydrogen bonding mechanisms such as holding water or for weak adhesive interactions. It is generally agreed that such hydroxyl groups are unlikely to enter into a polymerization reaction with urea-formaldehyde resins. The urea-nitrogen is simply a much better nucleophile than a sugar hydroxyl group under the curing conditions. It is for these reasons that the adhesive properties depended on the U-F resin and not the amount of N-sago or A-sago flour extender. Presumably, their amounts could become disruptive to the bonding process at some point, but finding that was not the objective of these experiments.

P-sago flour extender (Figure 4.7) does exhibit a slightly higher bond strength than N-sago or A-sago blended adhesive formulations. One might rationalize that observation as follows. Unlike A-sago or N-sago, P-sago has phosphate units at any of the substituted hydroxyl sites on the glucosyl extending units of the starch polymer chain (Figure 3.2). Solarek (1987) has noted that such phosphate moieties are linked at the C-6 position of the monomer glucocyl unit. However, some can be expected to be present also at C-2 and a few at C-3 as well. When such phosphate units replace the hydroxyl units, they give the glucosyl units a more polarizable moiety. One might expect the phosphate unit to give a stronger hydrogen bond. This may lead to slightly increased adhesive interaction. This is however somewhat speculative as the amount of phosphate is about 1% of the dry sago weight.

A P-sago adhesive is also slightly slower to cure than N-sago or A-sago adhesive formulations (Table 4.6). At the same time, the P-sago adhesive has a higher ultimate bond strength. It may be possible that the phosphate units are reacting to some extent with the U-F resin methylol groups. Such an interaction could slow the rate, but perhaps lead to a stronger bond. This suggests an area for future research and better understanding.

4.6 The Effects of Water vs. Solid Urea-Formaldehyde Content

In order to study the effect of water vs solid resin content, adhesive formulations were made where the relative proportions of urea-formaldehyde resin, catalyst and trial extenders were maintained constant while the percentage of water was varied. In effect this gave a series of formulations where the water/solid resin proportions varied inversely. The results are presented in Figure 4.8 and Figure 4.9 as a function of percent solid urea formaldehyde resin in the final adhesive mixture. The exact proportions of materials blended into these adhesive mixtures is shown in Table 3.2 for N-sago, Table 3.3 for A-sago, and Table 3.4 for P-sago flours.

Maximum bond strengths were found to be 10.3 MPa for N-sago extended adhesive, 10.6 MPa for the A-sago extended adhesive and 11.8 MPa for the P-sago extended adhesive formulation (Appendix 4.12).

Not surprising was the observation that the less the water or the more solid urea-formaldehyde resin the adhesive mixture contained, the higher was the curing rate. The cross-linking or curing reaction of a urea-formaldehyde resin is a condensation reaction between the $-NH_2$ moiety of a urea group and a methylol group. In this reaction water is liberated and when in excess, water can influence the rate of this reaction in a negative way.

Figure 4.8 shows that all of the trial extender adhesive formulations had a maximum curing rate at just over 40% solid urea-formaldehyde content. Higher concentrations of solid urea-formaldehyde resin were not practical and not tried in these experiments. The rates shown in Figure 4.8 are more than acceptable when compared to a standard urea-formaldehyde adhesive at 30% resin solids that was formulated with wheat flour. This standard resin had a curing rate of 0.027 MPa/sec. As one can see in Table 4.7 which summarizes these comparisons, P-sago and A-sago extended adhesive exhibit a curing rate of 0.022 and 0.030 MPa/sec., respectively, and N-sago extended adhesive showed a value of 0.035 MPa/sec.

Table 4.7 Comparison of Curing Rates of Trial Sago Flour Extenders to Standard Wheat Flour as an Extender

| Extenders | Curing rate MPa/sec. |
|---------------------|----------------------|
| Phosphorylated sago | 0.022 |
| Wheat flour | 0.027 |
| Acid modified sago | 0.030 |
| Natural sago | 0.035 |

- At 30% solid resin

Figure 4.9 compares the maximum bond strengths for N-sago, A-sago, and P-sago extended urea-formaldehyde formulations at the various percentages of solid resin also from 15 to just over 40% of the final adhesive mixture. As can be seen from Figure 4.9 increasing solid resin content results in increasing bond strength up to about 34 to 35%

solid resin and then the bond strength starts to decrease. All three extenders show this effect with their respective adhesive formulations. The peak for P-sago extended adhesive of 11.8 MPa occurs at 34.5 solid urea-formaldehyde resin. Peak strengths for N-sago (10.3 MPa) and A-sago (10.6 MPa) extended adhesives occurred at 35.4 and 35.9% resin solids respectively.

Table 4.8 Comparison of Maximum Bond Strengths of Sago Extenders to Wheat Flour Extender at 30 percent Solid Resin

| Extenders | Maximum bond strengths (MPa) |
|---------------------|------------------------------|
| Phosphorylated sago | 11.1 |
| Acid modified sago | 9.8 |
| Natural sago | 9.5 |
| Wheat flour | 8.9 |

Table 4.8 summarizes the peak bond strengths for the three types of sago extender and compares them to a standard urea-formaldehyde adhesive formulated with wheat flour. Compared to the "wheat flour standard" all of the sago extended adhesives had higher maximum bond strengths under these experimental conditions.

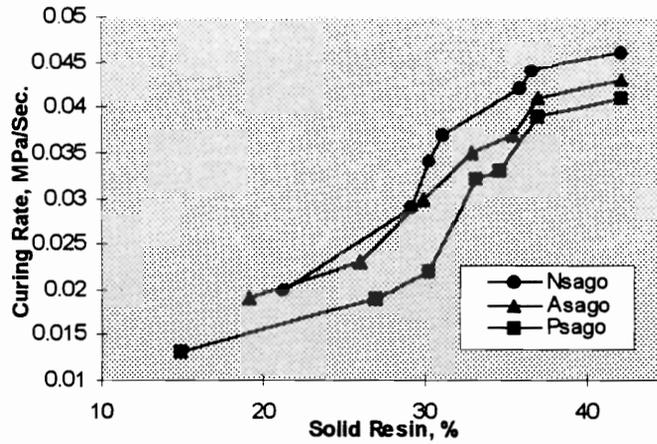


Figure 4.8. Curing rate of adhesive bonds extended with trial extenders at several water proportions.

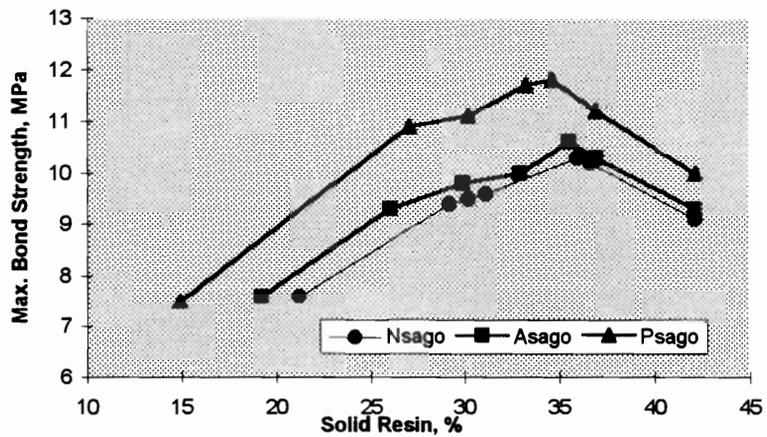


Figure 4.9. Maximum bond strength of adhesive bond extended with trial extenders at several water proportions.

Table 4.9 The Influence of Water Portion on Rate of Curing and Maximum Bond Strength

| Trial Extenders | Regressions Equation | R-squares | Max. Bond Strength, MPa* |
|----------------------------|-------------------------|-----------|-----------------------------|
| <u>Natural sago</u> | | | |
| 1 | $Y = -0.095 + 0.020 X$ | 85.4 | 7.6 |
| 2 | $Y = -0.052 + 0.029 X$ | 90.1 | 9.4 |
| 3 | $Y = -0.479 + 0.034 X$ | 86.1 | 9.5 |
| 4 | $Y = -0.722 + 0.037 X$ | 97.0 | 9.6 |
| 5 | $Y = -0.340 + 0.042 X$ | 97.0 | 10.3 |
| 6 | $Y = -1.022 + 0.044 X$ | 95.4 | 10.2 |
| 7 | $Y = -0.587 + 0.046 X$ | 85.1 | 9.1 |
| <u>Acid Modified Sago</u> | | | |
| 1 | $Y = -0.339 + 0.019 X$ | 80.9 | 7.6 |
| 2 | $Y = -0.281 + 0.023 X$ | 78.3 | 9.3 |
| 3 | $Y = -0.078 + 0.030 X$ | 71.4 | 9.8 |
| 4 | $Y = -0.461 + 0.035 X$ | 88.2 | 10.0 |
| 5 | $Y = -0.046 + 0.037 X$ | 10.2 | 10.6 |
| 6 | $Y = -0.041 + 0.041 X$ | 97.7 | 10.3 |
| 7 | $Y = -0.472 + 0.043 X$ | 89.3 | 9.3 |
| <u>Phosphorylated Sago</u> | | | |
| 1 | $Y = 0.286 + 0.013 X$ | 82.9 | 7.5 |
| 2 | $Y = 0.795 + 0.019 X$ | 75.5 | 10.9 |
| 3 | $Y = 0.370 + 0.022 X$ | 93.0 | 11.1 |
| 4 | $Y = 0.935 + 0.032 X$ | 92.3 | 11.7 |
| 5 | $Y = 0.400 + 0.033 X$ | 93.8 | 11.8 |
| 6 | $Y = -0.010 + 0.039 X$ | 85.5 | 11.2 |
| 7 | $Y = -0.097 + 0.041 X$ | 96.6 | 10.0 |

* Average of 5 replications
 Y = Maximum bond strength, MPa
 X = Curing time, seconds

Compared to the standard wheat flour extender any of the sago flour extenders had higher maximum bond strengths. At a level of 30% solid resin, wheat flour extended adhesive had an MBS of 8.9 MPa. While at the same solid resin level, N-sago, A-sago, and P-sago extended adhesives average MBS of 9.5, 9.8, and 11.1 MPa, respectively. The high MBS is similar with Nakazawa's work (Nakazawa et al. 1977 in Solarek 1986), who found that starch phosphate contributed to high adhesive strength for wood pieces, and more strength than rubber adhesives alone.

To get the same value in MBS (8.9 MPa) as wheat flour extender at 30% solid resin, P-sago, A-sago, and N-sago extender need about 21%, 24%, and 28% urea-formaldehyde solid resin, respectively. If the target of measure for adhesion is just bond strength, then P-sago extends UF adhesive is the best, A-sago the second, and N-sago the third quality. All are better than wheat flour. It should be noted here that bond strength is the basis of plywood quality control in the Japanese Agricultural Standard for plywood. This standard is used by Indonesia.

4.7 Temperature Effects

In order to study the effects of temperature variation, urea-formaldehyde adhesive mixtures were made with N-sago, A-sago, P-sago and wheat flour extenders in the formulations shown in Table 4.10.

Table 4.10 Adhesive Formulations to Test the Effects of Temperature Variation on Curing Rate

| Materials | Sago | | | Wheat flour |
|-----------|-------------------|---------------|----------|-------------|
| | Natural | Acid modified | Phosphor | |
| | grams | | | |
| UF-resin | 6.08 | 6.08 | 6.08 | 6.08 |
| Catalyst | 0.43 | 0.43 | 0.43 | 0.43 |
| Extender | 3.38 | 3.38 | 3.38 | 3.38 |
| Water | 0.27 | 0.41 | 0.68 | 1.15 |

In these formulations, the amount of urea-formaldehyde resin, catalyst, and extender were held constant. A temperature range of 84° C to 127° C was investigated.

The results are shown in Figure 4.10. Apriori, it was expected that each formulation would show a rise in curing rate as the temperature increased and perhaps at the higher temperatures a maximum rate plateau may have occurred.

However, each extender type showed an increasing curing rate with initial temperature rise, a rather sharp curing rate peak, and then a declining curing rate with farther increase in temperature.

More specifically, using P-sago as a trial extender to extend UF adhesive, the curing rate of the glue mixture rapidly increases if the temperature is increased up to 104° C (the actual/ real value is between 96-117° C). After this point continuing to increase temperature will rapidly decline the speed of curing of the glue mix which is added

with P-sago. N-sago and also A-sago like P-sago sharply increases in curing rate if the temperature is increased, but the maximum curing rate occurs at 96° C (between 85-104° C). After this, increasing the temperature the curing rate will rapidly decrease. The common extender yields the glue mix that has the curing rate slowly uncreases if the temperature increased up to 104° C, and then very slowly decreases (Figure 4.10).

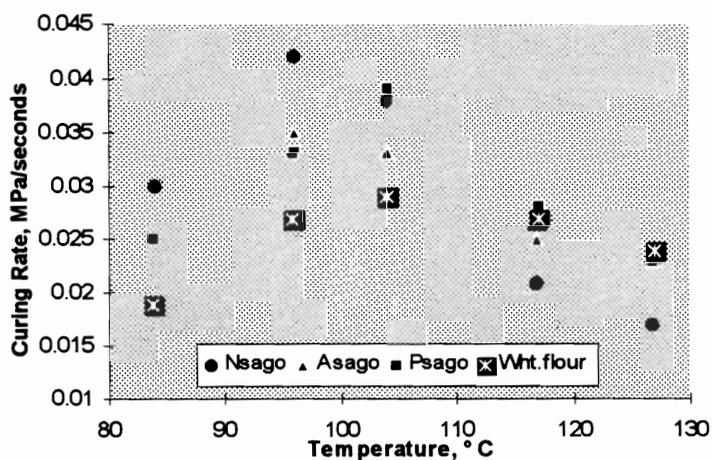


Figure 4.10. Curing rate of glue bond extended with trial extenders and wheat flour at several degrees of temperatures.

The speed of reaction increases when the temperature increases due to more energy available to react among the reactants. The trial extender phenomenon, however, is hard to explain. The interesting point is that they occur at slightly different temperatures (96 and 104° C.). At the top when the temperature continues to increase the rate of curing will in turn decline sharply. One possibility is that the formaldehyde at temperature near its boiling point (101° C) will boil and evaporate. Due to the consequent loss of formaldehyde content, less will be available in U-F polymer. Lower bond strength may result. The higher the temperature, the less formaldehyde, the lower the bonding strength may be. The second possible explanation might be linked to Wurzburgs' (1986) statement that when a starch is heated at some critical temperature, the hydrogen bonds of the granules weaken and they will imbibe water and swell greatly. Perhaps this occurs to A-sago and N-sago granules in the U-F adhesive, at 96° C., and for P-sago and wheat flour at 104° C. One could speculate that the swelling granule will hinder the polymerization of urea-formaldehyde, and therefore the curing rate will decrease. However, this is speculation at this point.

A third possibility is that a U-F resin when extended with these starchy extenders behaves as a thermosetting resin up to a certain temperature and then thermoplastic behavior sets in. The crystallinity of urea-formaldehyde resins is in question (Christiansen,1995). Urea-

formaldehyde adhesive as well as phenol-formaldehyde adhesive is a thermosetting resin, while cellulosic resin is thermoplastic (Maher,1960). Furthermore, Maher (1960) stated that an "alloy" or mix of thermosetting and thermoplastic materials may occur. One of the characters of thermoplastic resin is that when it is heated it will soften or melt at a certain higher temperature. In this experiment pulling the bond is done at the same temperature as the curing bond. In other words, testing the bonding strength is done at low to high temperatures that have been selected for the experiment's purpose.

The fourth possibility, according to Steiner (1987), is that before final cure occurs, the U-F resin behaves thermoplastically. The above information could be used to explain why the trial extenders when blended in a U-F resin, by increasing the temperature, the curing rate will be increased and at a certain level of temperature it will decreased, while the temperature continues to increase. (Figure 4.10).

At the high temperature the thermoplastic resin will melt, so the measurement of bond strength will appear "weaker" than if derived at the ambient temperature. To examine this possibility, additional research was done by collecting two sets of bond strength data. Two sets of samples were bonded under the same conditions (same mix ratio, temperature, and length of curing time), but the bond strengths were measured at different temperatures. The first

set was measured at the higher temperature, so that it was completed as usual by the ABES machine.

The second sample set was cured at the same temperature as the first set, but allowed to cool prior to pulling the bond apart. This cooling was done rapidly by applying cooling air to the bonds for ten seconds prior to pulling them apart. This was effective in decreasing sample temperature. Such data in the form of a curing rate ratio, ie hot pulled/cool pulled would indicate any thermoplastic behavior. The results are shown in Table 4.11.

The curing rates of N-sago are 0.042 of the cool test (CCR), and 0.017 of the hot test (HCR). If the HCR is divided by CCR (in percentage) then it is 40.86% (Table 4.8). This is called curing rate ratio (CRR). The CRR of P-sago and wheat flour are 33.77% and 55.81%. It can be predicted that U-F adhesive extending with trial extenders are more thermoplastic than one with wheat flour. If the glue mixture is a thermosetting so the HCR should be the same as CCR or CRR is equal or close to 100%. From the CRR value, the wheat flour made the U-F adhesive closer to 100% that means more thermosetting than N-sago and P-sago. Without the extender it is predicted that U-F adhesive will have a CRR value same or close to 100%.

Table 4.11 Cool and Hot Bonding Test of Wheat flour and Trial Extenders

| | Wheat Flour | Natural Sago | Phosphorylated Sago |
|-----------------------|----------------|-----------------|------------------------|
| Hot test | | | |
| Intercept | 3.2043 | 3.5739 | 2.9864 |
| Curing rate | 0.0240 | 0.0172 | 0.0232 |
| Corr. Coef. | 0.8820 | 0.8093 | 0.8195 |
| Std Error | 0.3036 | 0.3955 | 0.4122 |
| Cool test | | | |
| Intercept | 3.9564 | 3.8121 | 3.0321 |
| Curing rate | 0.0430 | 0.0421 | 0.0687 |
| Corr. Coef. | 0.8715 | 0.9287 | 0.8658 |
| Std Error | 0.2364 | 0.2778 | 0.2916 |
| CRH/CRC | 55.81 | 40.86 | 33.77 |
| (CRC-CRH) /CRC | 44.17 | 59.14 | 66.23 |

The percentage of all extenders added to the U-F glue mixtures were 92.65% based on the solid resin weight. All the extenders added were starch materials. Apparently the starch gave the U-F adhesive a thermoplastic component to this thermosetting adhesive.

The wheat flour has 15.5% protein (Robertson, 1977) while sago has no protein. Seemingly, that protein does not have a thermoplastic effect, so that U-F adhesive blends with the wheat flour is more thermosetting than if blend with N-sago flour.

P-sago has phosphate molecules, especially in the amylopectin part. Presumably, the phosphate may have thermoplastic tendency, therefore a U-F adhesive which blend with P-sago had the most thermoplastic than the others.

These results were unexpected and need to be further explored in future research to better define this anomalous observation about curing rates at higher temperatures.

V. SUMMARY AND CONCLUSIONS

5.1 Summary

Since Indonesia has banned its export of raw logs, plywood production has grown to such a rate that Indonesia is now the second largest producer of plywood in the world. The increasingly large volumes of urea-formaldehyde resins which are used in this plywood manufacturing also call for correspondingly larger amounts of wheat flour to be imported into Indonesia for use as an adhesive extender. In order to achieve less dependence on imported materials, it was deemed desirable to use local sources of starch for such extender purposes. However, earlier research had shown that simple substitution of sago starch flour for wheat flour was not satisfactory. Sago flour is an abundant and underutilized resource in Indonesia.

The goals of this research were to determine if alternative formulation strategies and/or modification of sago starch could be accomplished to make satisfactory urea-formaldehyde bonds using a sago starch flour. The guidelines employed in achieving these goals were that the adhesive mixture have a viscosity of about 1500 cPs and that satisfactory bonding would be achieved when the curing rate and maximum bond strength were comparable or better than a standard wheat flour extended urea-formaldehyde adhesive. Curing rates and maximum bond strengths would be measured using the "ABES" machine and meranti veneer.

Natural sago (unmodified), acid-modified sago and phosphorylated sago starch flours were used in this study. Direction for formulation was first achieved by exploring the interactive relationships of extender and water while holding the amount of urea-formaldehyde resin and catalyst constant. Viscosity was always a guiding principle in these experiments. From this information formulations were developed at 1500 cPs viscosity for N-sago, A-sago and P-sago extended urea-formaldehyde adhesives. These adhesive systems were studied using the ABES machine to obtain maximum curing rate and maximum bond strengths.

Formulations were developed that showed comparable curing rates to a wheat flour extended adhesive (0.027 MPa/s) for natural (0.035 MPa/s), acid modified (0.030 MPa/s), and phosphorylated (0.022 MPa/s) sago extended adhesives. All these adhesive mixtures had 30% resin solids and 1500 cPs viscosity. Also, maximum bond strengths compared favorably with wheat flour (8.9 MPa) extended adhesives; for natural (9.5 MPa), acid modified (9.8 MPa) and phosphorylated (11.1 MPa) sago extended adhesives. Also these values correspond to urea-formaldehyde formulations with 30% resin solids and 1500 cPs viscosity. The primary goals of this research were thus accomplished in that successful formulations were made using sago (modified and unmodified) flours to achieve curing rates and maximum bond strengths comparable to wheat flour extended adhesives. Since bond strength of plywood is a primary concern in the

Japanese Agricultural Standard for Plywood (which is used in Indonesia) it seems these results will be useful to the industry.

In one experiment where the effect of temperature on curing rate was studied using wheat flour and the sago flours as extenders, a significant anomalous and unexpected effect was observed. Quite unexpectedly, curing rates for all four types of extended urea-formaldehyde adhesives showed a rising rate of cure with resin temperatures up to about 94-104° C and then each sharply drops off with increasing temperature. Further studies where the bonds were cooled before pulling, rather than pulling them hot, suggests some sort of thermoplastic behavior. The author has no other good explanation for this observation.

5.2 Conclusions

In conclusion, this research has shown that sago starch can be used to formulate urea-formaldehyde adhesive for wood. The best one might be P-sago since its properties most closely resemble those of wheat flour.

5.3 Recommendations

Recommendations for future research are that the formulations developed in this research be tested to make actual plywood specimens. Adjustments may have to be made in

any scale-up process, but the principles outlined should hold up in the actual production of plywood using urea-formaldehyde extended sago starch products. The anomalous "thermoplastic" nature of the urea-formaldehyde curing process also needs to be further investigated.

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APPENDICES

Table 1 Physico-chemical Properties of Sago Starch and Wheat Flour

| Properties | Sago | Extender | Properties | Sago | Extender |
|---------------------------|--------------|--------------|--------------------|------------------|--------------|
| Moisture Contents, % | 14.06 | 12.52 | Weight/volume | | |
| | 15.91 | 11.58 | -loose | 0.640 | 0.423 |
| | <u>13.97</u> | <u>12.07</u> | | 0.670 | 0.425 |
| Average: | 14.25 | 12.06 | | <u>0.714</u> | <u>0.443</u> |
| $\sigma_{(n-1)}$: | 1.10 | 0.47 | Average: | 0.675 | 0.430 |
| | | | $\sigma_{(n-1)}$: | 0.037 | 0.011 |
| pH | 4.53 | 6.18 | -dense | 0.865 | 0.583 |
| | <u>4.93</u> | <u>5.90</u> | | 0.859 | 0.588 |
| Average: | 4.58 | 5.97 | | <u>0.845</u> | <u>0.579</u> |
| $\sigma_{(n-1)}$: | 0.33 | 0.19 | Average: | 0.856 | 0.583 |
| | | | $\sigma_{(n-1)}$: | 0.010 | 0.005 |
| Retrogradation, - minutes | 29 | 218 | Viscosity, cPs | 6100 | 8.4 |
| | 33 | 249 | | 6600 | 8.6 |
| | <u>32</u> | <u>230</u> | | <u>6900</u> | <u>8.8</u> |
| Average: | 31.3 | 232.3 | Average: | 6533 | 8.5 |
| $\sigma_{(n-1)}$: | 2.1 | 15.6 | $\sigma_{(n-1)}$: | 404 | 0.4 |
| - % volume | 52.6 | 79.2 | Weight/volume | 1.018 | 1.012 |
| | 50.0 | 77.9 | | 1.033 | 1.015 |
| | <u>53.8</u> | <u>80.8</u> | | <u>1.041</u> | <u>1.028</u> |
| Average: | 52.1 | 79.3 | Average: | 1.031 | 1.018 |
| $\sigma_{(n-1)}$: | 1.9 | 1.5 | $\sigma_{(n-1)}$: | 0.012 | 0.009 |
| Water intake: (g/g) (%) | 156.9 | 234.7 | | | |
| | 159.8 | 225.3 | | | |
| | <u>164.3</u> | <u>233.5</u> | | | |
| Average: | 160.3 | 231.2 | | | |
| $\sigma_{(n-1)}$: | 3.7 | 5.1 | | | |

Table 2 Physico-chemical Properties of Sago Starch
Treated by Soaking in 7.5 % of Hydrochloric Acid

| Properties | HCl Soaking Duration (days) | | | | | | EXTINDR |
|--------------------|-----------------------------|-------|-------|-------|-------|-------|---------|
| | 0 | 1 | 3 | 5 | 7 | 9 | |
| Moisture | 14.06 | 12.59 | 14.38 | 13.45 | 13.05 | 13.71 | 12.52 |
| Contents, % | 15.91 | 15.25 | 12.22 | 13.38 | 15.72 | 12.07 | 11.58 |
| | 13.97 | 15.01 | 14.52 | 13.08 | 14.23 | 14.12 | 12.07 |
| Average: | 14.65 | 14.28 | 13.71 | 13.30 | 14.33 | 13.30 | 12.06 |
| $\sigma_{(n-1)}$: | 1.10 | 1.47 | 1.29 | 0.19 | 1.34 | 1.08 | 0.47 |
| pH | 4.53 | 5.48 | 5.15 | 4.69 | 4.73 | 4.81 | 6.18 |
| | 4.27 | 4.63 | 4.74 | 4.38 | 4.76 | 5.15 | 5.82 |
| | 4.93 | 4.68 | 4.89 | 4.48 | 5.17 | 5.08 | 5.90 |
| Average: | 4.58 | 4.93 | 4.93 | 4.52 | 4.89 | 5.01 | 5.97 |
| $\sigma_{(n-1)}$: | 0.33 | 0.48 | 0.21 | 0.16 | 0.25 | 0.18 | 0.19 |
| Retrogradation, | | | | | | | |
| - minutes | 29 | 34 | 35 | 34 | 33 | 37 | 218 |
| | 33 | 38 | 34 | 44 | 43 | 38 | 249 |
| | 32 | 37 | 41 | 36 | 39 | 42 | 230 |
| Average: | 31.3 | 36.3 | 36.7 | 38.0 | 38.3 | 39.0 | 232 |
| $\sigma_{(n-1)}$: | 2.1 | 2.1 | 3.8 | 5.3 | 5.0 | 2.6 | 16 |
| - % volume | 52.6 | 50.0 | 60.0 | 57.7 | 56.4 | 53.8 | 79.2 |
| | 50.0 | 56.4 | 56.4 | 60.3 | 59.0 | 61.5 | 77.9 |
| | 53.8 | 52.6 | 55.1 | 56.4 | 61.5 | 62.8 | 80.8 |
| Average: | 52.1 | 53.0 | 57.2 | 58.1 | 59.0 | 59.4 | 79.3 |
| $\sigma_{(n-1)}$: | 1.9 | 3.2 | 2.5 | 2.0 | 2.6 | 4.9 | 1.5 |

Table 2 (Continued)

| Properties | HCl Soaking Duration (days) | | | | | | EXTNDR |
|--------------------|-----------------------------|-------|-------|-------|-------|-------|--------|
| | 0 | 1 | 3 | 5 | 7 | 9 | |
| Weight/volume | | | | | | | |
| - loose | 0.640 | 0.712 | 0.696 | 0.690 | 0.675 | 0.704 | 0.423 |
| | 0.670 | 0.731 | 0.675 | 0.675 | 0.690 | 0.686 | 0.425 |
| | 0.714 | 0.690 | 0.714 | 0.682 | 0.712 | 0.640 | 0.443 |
| Average: | 0.675 | 0.711 | 0.695 | 0.682 | 0.692 | 0.677 | 0.430 |
| $\sigma_{(n-1)}$: | 0.037 | 0.021 | 0.020 | 0.008 | 0.019 | 0.033 | 0.011 |
| - dense | 0.865 | 0.858 | 0.791 | 0.880 | 0.898 | 0.841 | 0.583 |
| | 0.859 | 0.867 | 0.831 | 0.849 | 0.819 | 0.833 | 0.588 |
| | 0.845 | 0.857 | 0.857 | 0.822 | 0.877 | 0.869 | 0.579 |
| Average: | 0.856 | 0.861 | 0.826 | 0.850 | 0.865 | 0.848 | 0.583 |
| $\sigma_{(n-1)}$: | 0.010 | 0.006 | 0.033 | 0.029 | 0.041 | 0.019 | 0.005 |
| Viscosity, cPs | | | | | | | |
| | 6100 | 8.9 | 3.5 | 2.5 | 2.6 | 1.9 | 8.4 |
| | 6600 | 6.0 | 3.0 | 2.5 | 2.0 | 2.0 | 8.6 |
| | 6900 | 5.9 | 2.8 | 2.9 | 2.3 | 2.5 | 8.8 |
| Average: | 6533 | 6.9 | 3.1 | 2.6 | 2.3 | 2.1 | 8.5 |
| $\sigma_{(n-1)}$: | 404 | 1.7 | 0.4 | 0.2 | 0.3 | 0.3 | 0.4 |
| Weight/volume | | | | | | | |
| | 1.018 | 1.026 | 1.015 | 1.017 | 1.016 | 1.010 | 1.012 |
| | 1.033 | 1.020 | 1.017 | 1.023 | 0.991 | 1.027 | 1.015 |
| | 1.041 | 1.017 | 1.029 | 1.019 | 1.030 | 1.021 | 1.028 |
| Average: | 1.031 | 1.021 | 1.020 | 1.020 | 1.012 | 1.019 | 1.018 |

Table 3 Phosphorylated Sago Properties From Natural and After One Day Acid Soaking

| Properties | Sago | Phosphorylated sago | | Wheat Flour |
|------------------------------|-----------|---------------------|--------------|-------------|
| | Untreated | Natural | Acid soaking | |
| Moisture Contents, % | 14.06 | 12.58 | 12.67 | 12.52 |
| | 13.97 | 14.12 | 13.20 | 11.58 |
| | 15.91 | 13.35 | 13.58 | 12.07 |
| Average: | 14.65 | 13.35 | 13.15 | 12.06 |
| $\sigma_{(n-1)}$: | 1.10 | 0.77 | 0.46 | 0.47 |
| pH | 4.53 | 7.08 | 5.87 | 6.18 |
| | 4.27 | 6.93 | 5.67 | 5.82 |
| | 4.95 | 6.60 | 6.01 | 5.90 |
| Average: | 4.58 | 6.87 | 5.85 | 5.97 |
| $\sigma_{(n-1)}$: | 0.34 | 0.25 | 0.17 | 0.19 |
| Retrogradation, - minutes | 29 | 64 | 129 | 218 |
| | 33 | 79 | 112 | 249 |
| | 32 | 71 | 124 | 230 |
| Average: | 31.3 | 71.3 | 121.7 | 232.3 |
| $\sigma_{(n-1)}$: | 2.1 | 7.5 | 8.7 | 15.6 |
| - volume, % | 52.6 | 61.9 | 73.0 | 79.2 |
| | 50.0 | 66.4 | 73.3 | 77.9 |
| | 53.8 | 61.8 | 76.4 | 80.8 |
| Average: | 52.1 | 63.4 | 74.1 | 79.3 |
| $\sigma_{(n-1)}$: | 1.9 | 2.6 | 6.0 | 1.5 |

Table 3 (Continued)

| Properties | Sago | Phosphorylated sago | | Wheat |
|--------------------|-----------|---------------------|--------------|-------|
| | Untreated | Natural | Acid soaking | Flour |
| Weight/Volume | | | | |
| - loose | .640 | .638 | .665 | .423 |
| | .670 | .577 | .673 | .425 |
| | .714 | .615 | .642 | .443 |
| Average: | .675 | .610 | .660 | .430 |
| $\sigma_{(n-1)}$: | .037 | .031 | .020 | .011 |
| - dense | .865 | .809 | .777 | .583 |
| | .859 | .793 | .748 | .588 |
| | .845 | .831 | .791 | .579 |
| Average: | .856 | .811 | .772 | .583 |
| $\sigma_{(n-1)}$: | .010 | .020 | .022 | .005 |
| Viscosity, cPs | 6100 | >10000 | 14.1 | 8.1 |
| | 6600 | >10000 | 12.3 | 8.6 |
| | 6900 | >10000 | 12.5 | 8.9 |
| Average: | 6533 | | 13.0 | 8.5 |
| $\sigma_{(n-1)}$: | 404 | | 1.0 | 0.4 |
| Water intake, % | 156.9 | 190.8 | 223.4 | 234.7 |
| | 159.8 | 201.1 | 231.7 | 225.3 |
| | 164.3 | 187.6 | 219.1 | 233.5 |
| Average: | 160.3 | 193.2 | 224.7 | 231.2 |
| $\sigma_{(n-1)}$: | 3.7 | 7.1 | 6.4 | 5.1 |

Note: Composition of 50 grams of Starch dissolved in 50 cc H₂O and 2.5 grams urea with 0.25 grams of NaH₂PO₄.H₂O and 0.25 grams of Na₂HPO₄

Table 4 Effect of Proportion of Natural Sago and Water on Viscosity of Adhesive Mixtures

| 94 % natural sago water, % | Viscosity cPs | 130 % natural sago water, % | Viscosity cPs | 182 % natural sago water, % | Viscosity cPs |
|----------------------------|---------------|-----------------------------|---------------|-----------------------------|---------------|
| 0 | 8700 | 26 | 2050 | 26 | 5500 |
| 13 | 2150 | 31 | 1300 | 34 | 2800 |
| 18 | 1100 | 34 | 1000 | 39 | 1650 |
| 20 | 1040 | 39 | 705 | 47 | 1100 |
| 26 | 800 | 47 | 525 | 52 | 860 |
| 39 | 400 | 60 | 300 | 60 | 640 |
| 47 | 290 | | | 78 | 370 |
| 52 | 200 | | | | |
| 72 | 133 | | | | |
| 78 | 115 | | | | |
| 200 | 15 | | | | |

* percentage base on solid resin

Table 5 Effect of Proportions of Acid Modified Sago and Water on Viscosity of Adhesive Mixtures

| 52 % Asago water, % | Viscosity cPs | 78 % Asago water, % | Viscosity cPs | 130 % Asago water, % | Viscosity cPs |
|---------------------|---------------|---------------------|---------------|----------------------|---------------|
| 0 | 2400 | 0 | 7300 | 26 | 4200 |
| 8 | 1320 | 13 | 2350 | 39 | 1750 |
| 18 | 720 | 18 | 1625 | 44 | 1200 |
| 26 | 500 | 26 | 1130 | 52 | 840 |
| 39 | 200 | 39 | 550 | 62 | 600 |
| | | 52 | 250 | 65 | 450 |
| | | 62 | 160 | 78 | 325 |
| | | 78 | 133 | 94 | 300 |
| | | 94 | 90 | | |

* percentage base on solid resin

Table 6 Effect of Proportions of Phosphorylated Sago and Water on Viscosity of Adhesive Mixtures

| 38 % Psago water, % | Viscosity cPs | 78 % Psago water, % | Viscosity cPs | 130 % Psago water, % | Viscosity cPs |
|---------------------------|------------------|---------------------------|------------------|----------------------------|------------------|
| 0 | 2700 | 26 | 2130 | 26 | 7200 |
| 13 | 1300 | 39 | 910 | 39 | 4200 |
| 26 | 550 | 52 | 420 | 52 | 1800 |
| 39 | 240 | 65 | 290 | 65 | 1100 |
| 52 | 120 | 78 | 200 | 78 | 800 |
| | | | | 91 | 480 |
| | | | | 104 | 325 |

* percentage base on solid resin

Table 7 Effect of Water Added on Viscosity of Straight Urea-Formaldehyde Adhesives

| Composition | | | | Viscosity (cPs) |
|-----------------|--------------------|-----------------|---------------|--------------------|
| Resin (gram) | Catalyst (gram) | Water (gram) | Water (%)* | |
| 6.4 | 0.4 | 0 | 0.0 | 530 |
| 6.4 | 0.4 | 0.7 | 18.2 | 180 |
| 6.4 | 0.4 | 1.0 | 26.0 | 113 |
| 6.4 | 0.4 | 1.8 | 46.9 | 60 |
| 6.4 | 0.4 | 2.0 | 52.1 | 55 |
| 6.4 | 0.4 | 2.3 | 59.9 | 42 |
| 6.4 | 0.4 | 3.0 | 78.1 | 30 |

*) Percent based on solid resin

Table 8 Moisture Content of Veneer as Specimens for Bonding Strength Test Measurement

| <u>weight of dry</u> | | Moisture content, % | <u>weight of dry</u> | | Moisture content, % |
|----------------------|-------|------------------------|----------------------|-------|------------------------|
| air | oven | | air | oven | |
| 1.969 | 1.855 | 6.15 | 1.797 | 1.686 | 6.58 |
| 1.834 | 1.711 | 7.17 | 1.740 | 1.634 | 6.49 |
| 1.869 | 1.761 | 6.13 | 1.815 | 1.712 | 6.12 |
| 1.803 | 1.699 | 6.12 | 1.911 | 1.794 | 6.52 |
| 1.830 | 1.716 | 6.64 | 1.898 | 1.785 | 6.33 |
| 1.944 | 1.831 | 6.13 | 1.874 | 1.762 | 6.36 |
| 1.990 | 1.857 | 7.16 | 1.755 | 1.638 | 7.14 |
| 1.763 | 1.649 | 6.91 | 1.869 | 1.750 | 6.80 |
| 1.879 | 1.757 | 6.94 | 1.829 | 1.723 | 6.15 |
| 1.825 | 1.708 | 6.85 | 1.707 | 1.599 | 6.75 |
| 1.830 | 1.726 | 6.03 | 1.777 | 1.671 | 6.34 |
| 1.693 | 1.584 | 6.88 | 1.794 | 1.691 | 6.09 |
| 1.830 | 1.707 | 7.21 | 1.719 | 1.609 | 6.84 |
| 1.759 | 1.651 | 6.54 | 1.768 | 1.649 | 7.22 |
| 1.854 | 1.725 | 6.86 | 1.800 | 1.698 | 6.01 |
| 1.850 | 1.743 | 6.14 | 1.835 | 1.717 | 6.83 |
| 1.936 | 1.810 | 6.96 | 1.781 | 1.675 | 6.33 |
| 1.779 | 1.665 | 6.84 | 1.762 | 1.649 | 6.85 |
| 1.812 | 1.698 | 6.71 | 1.854 | 1.732 | 7.04 |
| 1.799 | 1.687 | 6.64 | 1.839 | 1.721 | 6.86 |
| 1.868 | 1.746 | 6.99 | 1.861 | 1.742 | 6.83 |
| 1.790 | 1.675 | 6.87 | | | |
| 1.950 | 1.824 | 6.91 | | | |
| 1.814 | 1.703 | 6.52 | | | |
| 1.654 | 1.546 | 6.99 | | | |
| | | | Average: | | 6.65 |
| | | | $\sigma_{(n)}$ | : | 0.36 |

Table 9 The Bond Strength (in MPa) of UF-resin Adhesives Extended with Natural Sago Varied in Ratio of Materials/Water at Several Cure Time (in Seconds)

| Ratio of material (in grams) Viscosity keeps constant at 1500 cPs | | | | | | | | | | | | |
|--|-------|-------------|-------|------|-------|-------|-------|-------|-------|-------|----|-----|
| Materials | | Sample Code | | | | | | | | | | |
| | | N50R | | N83R | | N108R | | N133R | | N150R | | |
| CT | BS | CT | BS | CT | BS | CT | BS | CT | BS | CT | BS | |
| UF resin | | 6.1 | | 6.1 | | 6.1 | | 6.1 | | 6.1 | | 6.1 |
| Catalyst | | 0.4 | | 0.4 | | 0.4 | | 0.4 | | 0.4 | | 0.4 |
| Natural sago | | 1.82 | | 3.03 | | 3.94 | | 4.85 | | 5.47 | | |
| Water | | 0.13 | | 0.51 | | 0.79 | | 1.02 | | 1.23 | | |
| 40 | 0.147 | 60 | 0.647 | 40 | 0.367 | 80 | 2.091 | 50 | 1.437 | | | |
| 100 | 2.571 | 40 | 0.013 | 120 | 3.331 | 160 | 5.046 | 100 | 2.570 | | | |
| 60 | 1.282 | 40 | 0.048 | 80 | 2.214 | 40 | 0.474 | 150 | 3.793 | | | |
| 120 | 2.606 | 100 | 1.304 | 60 | 1.056 | 100 | 2.898 | 20 | 0.083 | | | |
| 80 | 2.055 | 20 | 0.024 | 100 | 3.026 | 20 | 0.042 | 130 | 3.846 | | | |
| 200 | 4.685 | 140 | 2.116 | 20 | 0.022 | 180 | 4.452 | 70 | 1.098 | | | |
| 200 | 5.022 | 80 | 1.007 | 160 | 5.009 | 60 | 1.266 | 120 | 2.466 | | | |
| 200 | 5.559 | 160 | 3.901 | 140 | 4.492 | 140 | 3.694 | 90 | 2.069 | | | |
| 300 | 6.227 | 180 | 4.521 | 90 | 2.313 | 120 | 3.922 | 40 | 0.403 | | | |
| 250 | 6.236 | 120 | 2.424 | 130 | 3.551 | 170 | 4.479 | 80 | 2.185 | | | |
| 150 | 5.022 | 150 | 3.658 | 70 | 1.563 | 150 | 4.492 | 165 | 4.309 | | | |
| 400 | 6.216 | 90 | 1.727 | 110 | 2.819 | 130 | 2.715 | 140 | 3.155 | | | |
| 130 | 3.114 | 145 | 2.745 | 150 | 5.150 | 130 | 3.158 | 180 | 4.489 | | | |
| 170 | 5.100 | 125 | 3.009 | 100 | 2.537 | 110 | 2.696 | 110 | 2.385 | | | |
| 220 | 6.241 | 115 | 2.595 | | | | | 210 | 5.698 | | | |
| 140 | 4.507 | 95 | 1.818 | | | | | | | | | |
| 180 | 4.367 | 130 | 2.591 | | | | | | | | | |
| 20 | 0.062 | | | | | | | | | | | |
| 50 | 0.814 | | | | | | | | | | | |
| 130 | 2.678 | | | | | | | | | | | |

Note: CT Curing time, in seconds
BS Bonding strength, in MPa

Table 10 The Bond Strength (in MPa) of UF-Adhesives Extended with Acid Modified Sago Varied in Ratio of Material/Water and Cure Times (in Seconds)

| Materials | | Sample Code | | | | | | | | | |
|---------------|-------|-------------|-------|------|-------|------|-------|-------|-------|-------|----|
| | | A50R | | A67R | | A83R | | A108R | | A125R | |
| CT | BS | CT | BS | CT | BS | CT | BS | CT | BS | CT | BS |
| UF resin | | 6.1 | | 6.1 | | 6.1 | | 6.1 | | 6.1 | |
| Catalyst | | 0.4 | | 0.4 | | 0.4 | | 0.4 | | 0.4 | |
| Acid Md. sago | | 1.82 | | 2.44 | | 3.03 | | 3.94 | | 4.56 | |
| Water | | 0.15 | | 0.40 | | 0.66 | | 1.03 | | 1.34 | |
| 80 | 1.812 | 80 | 3.073 | 110 | 2.245 | 80 | 0.057 | 60 | 1.684 | | |
| 160 | 3.417 | 150 | 4.486 | 100 | 2.385 | 30 | 0.766 | 130 | 3.834 | | |
| 40 | 0.224 | 20 | 0.044 | 20 | 0.029 | 80 | 2.313 | 160 | 5.218 | | |
| 100 | 2.511 | 60 | 1.696 | 60 | 1.508 | 130 | 3.406 | 70 | 1.999 | | |
| 20 | 0.002 | 130 | 3.578 | 120 | 2.820 | 40 | 0.791 | 100 | 2.971 | | |
| 120 | 3.369 | 50 | 1.547 | 30 | 0.029 | 150 | 4.482 | 140 | 4.388 | | |
| 70 | 1.252 | 120 | 3.829 | 50 | 0.950 | 20 | 0.051 | 120 | 2.817 | | |
| 130 | 1.707 | 30 | 0.073 | 150 | 3.431 | 100 | 2.635 | 150 | 5.008 | | |
| 50 | 0.858 | 70 | 2.185 | 80 | 1.476 | 70 | 1.901 | 90 | 2.161 | | |
| 90 | 2.258 | 40 | 1.154 | 130 | 3.486 | 90 | 2.810 | 110 | 2.457 | | |
| 110 | 2.308 | 160 | 5.599 | 160 | 4.936 | 160 | 4.577 | 30 | 0.159 | | |
| 60 | 1.218 | 90 | 2.632 | 90 | 2.193 | 50 | 1.158 | 50 | 1.106 | | |
| 115 | 2.916 | 100 | 2.969 | 140 | 3.215 | 110 | 2.629 | 40 | 0.629 | | |
| 110 | 2.180 | 110 | 3.801 | 70 | 1.893 | 120 | 3.701 | 80 | 1.634 | | |
| 105 | 2.433 | 140 | 4.034 | 40 | 0.684 | 140 | 3.825 | 20 | 0.059 | | |
| 30 | 0.059 | | | 115 | 2.998 | 60 | 1.633 | 105 | 2.368 | | |
| 45 | 0.429 | | | 155 | 4.530 | | | 85 | 1.656 | | |
| 75 | 1.464 | | | | | | | 95 | 1.734 | | |
| 110 | 2.178 | | | | | | | | | | |
| 110 | 2.224 | | | | | | | | | | |
| 113 | 2.522 | | | | | | | | | | |
| 65 | 1.347 | | | | | | | | | | |

Note: CT Curing time, in seconds
BS Bonding strength in MPa

Table 11 The Bond Strength (in MPa) of UF-Adhesives
 Extended with Phosphorylated Sago Varied in Ratio of
 Materials/Water and Cure Times (in Seconds)

Ratio of material (in grams)

Viscosity keeps constant at 1500 cPs

| Materials | Sample Code | | | | | | | | | |
|-----------|-------------|-------|------|-------|------|-------|-------|-------|-------|-------|
| | P34R | | P58R | | P83R | | P108R | | P133R | |
| | CT | BS | CT | BS | CT | BS | CT | BS | CT | BS |
| UF resin | 6.1 | | 6.1 | | 6.1 | | 6.1 | | 6.1 | |
| Catalyst | 0.4 | | 0.4 | | 0.4 | | 0.4 | | 0.4 | |
| Psago | 1.24 | | 1.83 | | 3.03 | | 3.94 | | 4.85 | |
| Water | 0.27 | | 0.74 | | 0.87 | | 1.02 | | 1.26 | |
| | 50 | 0.339 | 70 | 1.258 | 90 | 2.430 | 110 | 2.309 | 160 | 3.797 |
| | 20 | 0.005 | 120 | 2.247 | 40 | 0.840 | 150 | 2.687 | 70 | 1.786 |
| | 140 | 3.489 | 60 | 0.929 | 20 | 0.039 | 20 | 0.029 | 80 | 1.751 |
| | 100 | 1.763 | 150 | 2.988 | 60 | 1.407 | 60 | 1.407 | 90 | 1.961 |
| | 80 | 1.771 | 50 | 0.670 | 130 | 2.954 | 140 | 3.015 | 155 | 3.087 |
| | 150 | 2.526 | 170 | 3.251 | 140 | 2.750 | 50 | 1.077 | 60 | 1.565 |
| | 60 | 1.381 | 20 | 0.018 | 70 | 1.664 | 40 | 0.716 | 130 | 2.264 |
| | 30 | 0.046 | 40 | 0.744 | 160 | 3.359 | 120 | 2.524 | 30 | 0.086 |
| | 70 | 1.295 | 160 | 3.396 | 120 | 5.724 | 160 | 3.542 | 150 | 2.485 |
| | 120 | 2.352 | 90 | 1.932 | 80 | 2.041 | 90 | 1.937 | 140 | 2.925 |
| | 160 | 3.419 | 190 | 2.595 | 110 | 2.452 | 80 | 1.887 | 20 | 0.048 |
| | 90 | 2.080 | 115 | 1.978 | 100 | 2.751 | 30 | 0.227 | 50 | 1.353 |
| | 40 | 0.582 | 80 | 1.272 | 50 | 1.221 | 130 | 2.642 | 40 | 0.603 |
| | 130 | 2.847 | 135 | 2.932 | 150 | 3.521 | 70 | 1.665 | 110 | 2.200 |
| | 110 | 2.147 | 100 | 1.961 | 30 | 0.197 | 100 | 2.447 | 120 | 2.442 |
| | | | | | 105 | 2.176 | 155 | 3.650 | 100 | 2.354 |
| | | | | | 115 | 2.795 | | | 45 | 0.966 |

Note: CT Curing time, in seconds
 BS Bonding strength, in MPa

Table 12 Maximum Bond Strength of Adhesive Mixtures Blended with Several Different Percentages of Trial Extenders (in MPa)

| Sample Code | Replications | | | | Average | $\sigma_{(n-1)}$ |
|----------------------------|--------------|-------|-------|-------|---------|------------------|
| | 1 | 2 | 3 | 4 | | |
| Natural sago | | | | | | |
| N50R | 9.79 | 9.90 | 10.73 | | 10.14 | 0.51 |
| N83R | 10.09 | 9.90 | 10.28 | | 10.09 | 0.19 |
| N108R | 10.25 | 10.58 | 9.68 | | 10.17 | 0.46 |
| N133R | 8.70 | 9.79 | 8.89 | | 9.13 | 0.58 |
| N150R | 8.70 | 11.29 | 9.23 | | 9.74 | 1.37 |
| Acid modified sago | | | | | | |
| A50R | 10.80 | 10.65 | 10.43 | | 10.63 | 0.19 |
| A67R | 12.78 | 11.85 | 11.75 | | 12.13 | 0.57 |
| A83R | 10.40 | 9.35 | 11.10 | 10.68 | 10.38 | 0.75 |
| A108R | 10.23 | 9.93 | 10.35 | | 10.17 | 0.22 |
| A125R | 10.19 | 9.72 | 10.85 | 10.09 | 10.25 | 0.57 |
| Phosphorylated sago | | | | | | |
| P34R | 11.58 | 11.07 | 9.33 | 10.16 | 10.54 | 1.00 |
| P58R | 11.52 | 11.10 | 10.20 | | 10.94 | 0.67 |
| P83R | 11.93 | 10.58 | 10.28 | | 10.93 | 0.88 |
| P108R | 11.93 | 11.29 | 10.43 | 11.70 | 11.34 | 0.66 |
| P133R | 12.75 | 11.67 | 11.88 | 12.18 | 12.12 | 0.47 |

Table 13 Bond Strength of Natural Sago Extended U-F Adhesives at Several Water Percentages, in MPa.

| Ratio of materials, in grams | | | | | | |
|------------------------------|-------------|------|------|------|------|------|
| Materials | Sample Code | | | | | |
| | N21V | N29V | N30V | N31V | N36V | N37V |
| UF-resin | 6.08 | 6.08 | 6.08 | 6.08 | 6.08 | 6.08 |
| Catalyst | 0.43 | 0.43 | 0.43 | 0.43 | 0.43 | 0.43 |
| Natural sago | 3.38 | 3.38 | 3.38 | 3.38 | 3.38 | 3.38 |
| Water | 7.29 | 2.62 | 2.18 | 1.83 | 0.27 | 0.00 |

| N21V | | N29V | | N30V | | N31V | | N36V | | N37V | |
|------|-------|------|-------|------|-------|------|-------|------|-------|------|-------|
| CT | BS |
| 40 | 0.940 | 60 | 2.236 | 100 | 2.508 | 80 | 2.702 | 100 | 4.513 | 60 | 1.802 |
| 80 | 1.574 | 80 | 2.921 | 130 | 4.347 | 40 | 0.445 | 120 | 4.429 | 120 | 4.509 |
| 140 | 2.126 | 100 | 2.956 | 160 | 4.899 | 120 | 3.530 | 140 | 5.592 | 160 | 6.810 |
| 100 | 1.676 | 140 | 5.055 | 140 | 3.647 | 160 | 4.995 | 160 | 5.016 | 140 | 5.533 |
| 160 | 3.743 | 160 | 3.214 | 120 | 2.818 | 140 | 4.541 | 150 | 6.262 | 100 | 3.438 |
| 160 | 3.524 | 130 | 3.393 | 90 | 2.564 | 100 | 2.783 | 130 | 5.027 | 80 | 2.683 |
| 130 | 2.196 | 90 | 4.250 | 80 | 1.568 | 60 | 1.386 | 110 | 4.503 | 40 | 0.772 |
| 150 | 4.313 | 50 | 1.413 | 60 | 1.474 | 20 | 0.071 | 90 | 3.061 | 20 | 0.038 |
| 120 | 1.576 | 20 | 0.029 | 40 | 0.576 | 70 | 2.184 | 70 | 2.101 | 30 | 0.071 |
| 60 | 1.015 | 30 | 0.666 | 20 | 0.035 | 50 | 1.193 | 50 | 1.773 | 70 | 2.035 |
| 90 | 1.441 | 70 | 2.097 | 30 | 0.543 | 90 | 2.250 | 30 | 1.137 | 110 | 3.776 |
| 145 | 1.855 | 90 | 2.899 | 50 | 1.172 | 110 | 4.013 | 20 | 0.639 | 150 | 4.485 |
| 170 | 2.185 | 110 | 2.903 | 70 | 2.996 | 130 | 4.498 | | | 130 | 4.499 |
| 150 | 2.183 | 40 | 1.319 | 110 | 4.487 | 90 | 2.924 | | | 90 | 2.495 |
| 160 | 2.909 | 120 | 3.298 | | | | | | | | |
| 155 | 2.675 | 160 | 4.553 | | | | | | | | |
| 125 | 3.115 | 140 | 3.589 | | | | | | | | |
| 115 | 2.181 | 100 | 2.626 | | | | | | | | |
| 70 | 1.044 | 55 | 1.352 | | | | | | | | |
| 110 | 1.467 | 125 | 3.266 | | | | | | | | |
| 20 | 0.367 | | | | | | | | | | |
| 50 | 0.939 | | | | | | | | | | |
| 125 | 2.952 | | | | | | | | | | |
| 170 | 3.468 | | | | | | | | | | |
| 75 | 1.713 | | | | | | | | | | |
| 100 | 1.696 | | | | | | | | | | |
| 30 | 0.564 | | | | | | | | | | |
| 55 | 1.213 | | | | | | | | | | |
| 125 | 2.480 | | | | | | | | | | |
| 115 | 1.824 | | | | | | | | | | |
| 120 | 1.550 | | | | | | | | | | |
| 110 | 1.874 | | | | | | | | | | |

Note: CT Cure time, in seconds
BS Bond strength, in MPa

Table 14 Bond Strength of Acid Modified Sago Extended U-F Adhesives at Several Water Percentages, in MPa.

Ratio of materials, in grams

| Materials | Sample Code | | | | | |
|---------------|-------------|------|------|------|------|------|
| | A19V | A26V | A30V | A33V | A36V | A37V |
| UF-resin | 6.08 | 6.08 | 6.08 | 6.08 | 6.08 | 6.08 |
| Catalyst | 0.43 | 0.43 | 0.43 | 0.43 | 0.43 | 0.43 |
| Acid mod sago | 3.38 | 3.38 | 3.38 | 3.38 | 3.38 | 3.38 |
| Water | 9.11 | 4.13 | 2.31 | 1.21 | 0.38 | 0.00 |

| A19V | | A26V | | A30V | | A33V | | A36V | | A37V | |
|------|-------|------|-------|------|-------|------|-------|------|-------|------|-------|
| CT | BS |
| 100 | 2.654 | 100 | 2.384 | 100 | 3.793 | 100 | 4.323 | 100 | 4.183 | 100 | 5.077 |
| 120 | 2.606 | 120 | 2.419 | 120 | 2.381 | 120 | 4.544 | 140 | 5.074 | 130 | 6.228 |
| 140 | 2.112 | 140 | 4.174 | 140 | 4.474 | 140 | 4.454 | 160 | 6.188 | 110 | 5.079 |
| 160 | 3.871 | 160 | 5.037 | 160 | 4.516 | 160 | 6.198 | 120 | 3.562 | 90 | 4.942 |
| 130 | 2.843 | 130 | 2.917 | 130 | 4.407 | 130 | 5.599 | 80 | 2.426 | 70 | 4.502 |
| 110 | 1.912 | 110 | 2.525 | 110 | 2.981 | 110 | 3.573 | 60 | 2.580 | 50 | 2.299 |
| 90 | 1.897 | 90 | 2.137 | 90 | 4.458 | 90 | 4.766 | 40 | 1.326 | 30 | 1.152 |
| 70 | 1.947 | 70 | 2.610 | 70 | 3.614 | 70 | 3.066 | 20 | 0.101 | 20 | 0.812 |
| 30 | 0.786 | 50 | 1.349 | 50 | 2.015 | 50 | 2.105 | 50 | 1.739 | 60 | 2.564 |
| 20 | 0.415 | 30 | 1.289 | 30 | 1.039 | 30 | 1.838 | 70 | 2.711 | 80 | 3.512 |
| 40 | 1.031 | 20 | 0.442 | 40 | 1.493 | 20 | 0.878 | 90 | 4.381 | | |
| 60 | 1.818 | 40 | 1.353 | 60 | 2.057 | 40 | 1.461 | 110 | 4.231 | | |
| 50 | 1.366 | 60 | 1.766 | 80 | 2.979 | 60 | 2.732 | 150 | 4.535 | | |
| 80 | 2.255 | 80 | 2.307 | 95 | 3.782 | 80 | 2.979 | 130 | 5.051 | | |
| 85 | 1.695 | 135 | 2.453 | 20 | 0.610 | 85 | 2.989 | 30 | 1.037 | | |
| | | | | 85 | 4.006 | | | | | | |
| | | | | 65 | 2.908 | | | | | | |
| | | | | 100 | 4.563 | | | | | | |
| | | | | 105 | 3.738 | | | | | | |
| | | | | 115 | 4.784 | | | | | | |
| | | | | 110 | 5.155 | | | | | | |

Note: CT Cure time, in seconds
BS Bond strength, in MPa

Table 15 Bond Strength of Phosphorylated Sago Extended U-F Adhesives at Several Water Percentages, in MPa.

| Ratio of materials, in grams | | | | | | | | | | | | |
|------------------------------|-------------|--|------|--|------|--|------|--|------|--|------|--|
| Materials | Sample Code | | | | | | | | | | | |
| | P15V | | P27V | | P30V | | P33V | | P35V | | P37V | |
| UF-resin | 6.08 | | 6.08 | | 6.08 | | 6.08 | | 6.08 | | 6.08 | |
| Catalyst | 0.43 | | 0.43 | | 0.43 | | 0.43 | | 0.43 | | 0.43 | |
| Acid mod sago | 3.38 | | 3.38 | | 3.38 | | 3.38 | | 3.38 | | 3.38 | |
| Water | 14.57 | | 3.64 | | 2.19 | | 1.09 | | 0.66 | | 0.00 | |

| P15V | | P27V | | P30V | | P33V | | P35V | | P37V | |
|------|-------|------|-------|------|-------|------|-------|------|-------|------|-------|
| CT | BS |
| 100 | 1.104 | 100 | 2.283 | 100 | 2.503 | 100 | 4.312 | 100 | 3.709 | 110 | 3.009 |
| 140 | 1.937 | 140 | 3.233 | 120 | 2.839 | 120 | 5.050 | 120 | 4.525 | 130 | 5.119 |
| 170 | 2.539 | 160 | 2.873 | 140 | 3.902 | 140 | 4.986 | 140 | 5.391 | 150 | 6.851 |
| 190 | 2.579 | 150 | 4.430 | 160 | 2.442 | 130 | 5.621 | 160 | 4.989 | 120 | 3.581 |
| 160 | 2.061 | 130 | 4.551 | 150 | 3.741 | 110 | 5.627 | 130 | 4.508 | 100 | 4.323 |
| 130 | 2.441 | 120 | 3.346 | 130 | 2.362 | 90 | 4.521 | 110 | 3.532 | 80 | 2.741 |
| 110 | 1.354 | 90 | 3.049 | 110 | 2.160 | 70 | 3.636 | 90 | 3.302 | 60 | 2.599 |
| 90 | 1.366 | 70 | 2.040 | 90 | 2.047 | 50 | 2.377 | 70 | 1.956 | 40 | 1.801 |
| 70 | 1.305 | 50 | 1.305 | 70 | 1.927 | 30 | 1.647 | 50 | 1.923 | 20 | 0.898 |
| 50 | 0.959 | 30 | 1.686 | 50 | 1.648 | 20 | 1.267 | 30 | 1.833 | 90 | 3.647 |
| 30 | 0.728 | 20 | 0.651 | 30 | 1.071 | 40 | 2.073 | 20 | 0.955 | 140 | 5.586 |
| 20 | 0.357 | 40 | 1.511 | 20 | 0.752 | 60 | 3.080 | 60 | 2.460 | | |
| 120 | 2.372 | 60 | 2.428 | 40 | 1.312 | 80 | 3.685 | 80 | 3.205 | | |
| 105 | 1.945 | 80 | 2.379 | 60 | 1.881 | | | 40 | 1.842 | | |
| 125 | 3.225 | | | 80 | 2.139 | | | | | | |

Note: CT Cure time, in seconds
BS Bond strength, in MPa

Table 16 Bond Strength of Evaporated U-F Adhesive Extends with Trial Sago at Several Water Percentages, in MPa

| Materials (grams) | Sample Code | | | | | |
|----------------------|-------------|-------|------|-------|------|-------|
| | N42V | | A42V | | P42V | |
| U-F resin* | 6.08 | | 6.08 | | 6.08 | |
| Catalyst | 0.42 | | 0.42 | | 0.42 | |
| Trial sago | 3.40 | | 3.40 | | 3.40 | |
| Water | 0.96 | | 0.96 | | 0.96 | |
| | CT | BS | CT | BS | CT | BS |
| | 100 | 3.198 | 100 | 4.868 | 100 | 4.205 |
| | 120 | 5.593 | 120 | 4.543 | 120 | 4.468 |
| | 140 | 5.616 | 140 | 5.057 | 140 | 5.613 |
| | 130 | 5.589 | 150 | 7.328 | 160 | 6.233 |
| | 110 | 4.480 | 130 | 4.526 | 150 | 6.251 |
| | 90 | 2.654 | 110 | 4.539 | 130 | 6.783 |
| | 70 | 2.184 | 90 | 4.474 | 110 | 4.510 |
| | 50 | 2.104 | 70 | 2.173 | 90 | 3.843 |
| | 30 | 0.274 | 50 | 2.011 | 70 | 2.976 |
| | 20 | 0.028 | 30 | 0.530 | 50 | 2.684 |
| | 60 | 2.226 | 20 | 0.025 | 30 | 1.027 |
| | 80 | 4.478 | 80 | 2.637 | 20 | 0.052 |
| | 105 | 3.677 | | | | |
| | 40 | 1.344 | | | | |
| | 75 | 4.328 | | | | |

Note: * evaporated UF adhesive, solid resin = 75%
 CT Cure time, in seconds
 BS Bonding strength, in MPa

Table 17 Maximum Bond Strength of UF Adhesive Blended With
Trial Extenders at Several Percentage of Solid
Resin, in MPa

| Natural sago | | | | | | | |
|----------------------------|------|------|------|------|------|------|------|
| Solid resin, % | 21.2 | 29.2 | 30.2 | 31.1 | 35.9 | 36.6 | 42.1 |
| Sample Code | N21V | N29V | N30V | N31V | N36V | N37V | N42V |
| | 6.6 | 9.4 | wb | 9.9 | 10.1 | 10.6 | 8.6 |
| | 7.8 | 8.9 | 9.1 | 8.9 | 10.3 | 9.1 | 8.7 |
| | 8.6 | 9.9 | 9.4 | 9.1 | 9.6 | 11.1 | 8.4 |
| | 8.1 | 9.0 | 9.5 | 10.2 | 11.2 | 9.4 | 9.1 |
| | 6.7 | 9.8 | 10.1 | 9.9 | 10.5 | 10.7 | 11.7 |
| Average | 7.6 | 9.4 | 9.5 | 9.6 | 10.3 | 10.2 | 9.3 |
| $\sigma_{(n-1)}$ | .88 | .45 | .42 | .57 | .59 | 0.88 | 1.37 |
| Acid Modified sago | | | | | | | |
| Solid resin, % | 19.2 | 26.0 | 29.9 | 32.9 | 35.5 | 36.9 | 42.1 |
| Sample Code | A19V | A26V | A30V | A33V | A36V | A37V | A42V |
| | 7.8 | 8.3 | 9.7 | 10.6 | 10.6 | 10.1 | 10.7 |
| | 8.4 | 9.9 | 8.7 | 8.6 | 9.4 | 9.2 | 7.7 |
| | 7.9 | 9.0 | 10.5 | 10.7 | 11.5 | 10.9 | 9.9 |
| | 7.1 | 10.5 | 8.9 | wb | 10.9 | 11.2 | 10.6 |
| | 7.0 | 8.8 | 11.3 | 10.2 | 10.4 | wb | 7.5 |
| Average | 7.6 | 9.3 | 9.8 | 10.0 | 10.6 | 10.4 | 9.3 |
| $\sigma_{(n-1)}$ | .59 | .89 | 1.01 | .97 | .77 | .90 | 1.56 |
| Phosphorylated sago | | | | | | | |
| Solid resin, % | 14.9 | 27.0 | 30.2 | 33.2 | 34.6 | 36.9 | 42.1 |
| Sample Code | P15V | P27V | P30V | P33V | P35V | P37V | P42V |
| | 7.1 | 10.2 | 10.6 | 12.8 | 12.6 | 11.6 | 10.2 |
| | 7.5 | 10.2 | 10.5 | 11.3 | 12.1 | 8.8 | 11.0 |
| | 8.2 | 11.9 | 11.6 | 9.8 | 10.8 | 11.4 | 9.5 |
| | 6.4 | 10.9 | 10.6 | 12.3 | 11.7 | 12.1 | 8.6 |
| | 8.4 | 11.1 | 12.0 | 12.4 | wb | 12.2 | 10.5 |
| Average | 7.6 | 10.9 | 11.1 | 11.7 | 11.8 | 11.2 | 10.0 |
| $\sigma_{(n-1)}$ | .87 | .71 | .69 | 1.21 | 0.76 | 1.39 | .93 |

31.8% Wheat flour: 8.9, 8.2, 11.3, 8.6, and 9.6 MPa

Average: 9.3 MPa; $\sigma_{(n-1)}$: 1.22

Pressing temperature: 95° C for 400 seconds

Table 18 Bond Strength of U-F Adhesive Extended with Natural Sago at Several Temperatures, in MPa

| Material grams | UF resin 6.1 | | Catalyst 0.43 | | Natural sago 3.38 | | Water 0.27 | | | |
|----------------|-----------------|-----|------------------|-----|----------------------|-----|---------------|-----|-------|----|
| Temp. °C. | 84 | | 96 | | 104 | | 117 | | 127 | |
| Sample Code | N85T | | N95T | | N105T | | N115T | | N125T | |
| | CT | BS | CT | BS | CT | BS | CT | BS | CT | BS |
| 100 | 1.730 | 100 | 4.513 | 90 | 4.425 | 80 | 3.875 | 80 | 4.494 | |
| 130 | 2.532 | 120 | 4.429 | 120 | 4.982 | 120 | 5.607 | 100 | 5.097 | |
| 160 | 3.778 | 140 | 5.592 | 150 | 4.921 | 130 | 6.195 | 110 | 5.086 | |
| 180 | 4.332 | 160 | 5.016 | 130 | 5.583 | 100 | 3.987 | 60 | 4.380 | |
| 140 | 2.552 | 150 | 6.262 | 110 | 4.437 | 70 | 5.019 | 40 | 4.510 | |
| 110 | 1.933 | 130 | 5.027 | 70 | 4.409 | 50 | 4.773 | 20 | 3.327 | |
| 80 | 1.461 | 110 | 4.503 | 50 | 2.738 | 20 | 2.273 | 15 | 2.550 | |
| 60 | 0.669 | 90 | 3.061 | 30 | 1.233 | 15 | 1.253 | 30 | 4.296 | |
| 40 | 0.103 | 70 | 2.101 | 20 | 0.570 | 35 | 3.792 | 50 | 3.761 | |
| 150 | 2.304 | 50 | 1.773 | 40 | 1.950 | 110 | 4.528 | 70 | 6.712 | |
| 155 | 4.481 | 30 | 1.137 | 60 | 3.175 | 90 | 5.074 | 90 | 5.543 | |
| 20 | 0.042 | 20 | 0.639 | 80 | 3.883 | 105 | 5.179 | 25 | 4.317 | |
| | | | | | | 60 | 4.429 | 70 | 5.040 | |
| | | | | | | 25 | 2.572 | | | |

Note: CT Cure time, in seconds
BS Bond strength, in MPa

Table 19 Bond Strength of U-F Adhesive Extended with Acid Modified Sago at Several Temperatures, in MPa

| Material: | UF resin | Catalyst | Acid Mod. sago | Water |
|-----------|----------|----------|----------------|-------|
| grams : | 6.1 | 0.43 | 3.38 | 0.38 |

| Temp. °C. | 84 | | 96 | | 104 | | 117 | | 127 | |
|-------------|------|-------|------|-------|-------|-------|-------|-------|-------|--------|
| Sample Code | A85T | | A95T | | A105T | | A115T | | A125T | |
| | CT | BS | CT | BS | CT | BS | CT | BS | CT | BS |
| | 100 | 2.285 | 100 | 4.183 | 90 | 4.051 | 80 | 4.629 | 80 | 4.295 |
| | 130 | 2.523 | 140 | 5.074 | 120 | 5.062 | 110 | 4.468 | 100 | 5.424 |
| | 170 | 3.577 | 160 | 6.188 | 150 | 4.506 | 140 | 5.680 | 70 | 4.959 |
| | 190 | 4.511 | 120 | 3.562 | 100 | 4.473 | 160 | 4.911 | 50 | 4.460 |
| | 160 | 3.192 | 80 | 2.426 | 80 | 4.492 | 130 | 5.075 | 30 | 3.693 |
| | 140 | 2.943 | 60 | 2.580 | 60 | 3.754 | 100 | 5.053 | 20 | 3.3243 |
| | 80 | 1.568 | 40 | 1.326 | 40 | 2.284 | 70 | 4.447 | 15 | 2.670 |
| | 60 | 1.115 | 20 | 0.101 | 30 | 1.224 | 40 | 3.673 | 90 | 4.464 |
| | 40 | 0.076 | 50 | 1.739 | 20 | 0.424 | 20 | 2.269 | 75 | 5.511 |
| | 110 | 2.780 | 70 | 2.711 | 30 | 1.224 | 15 | 1.799 | 95 | 5.5671 |
| | | | 90 | 4.381 | 50 | 2.628 | 30 | 3.261 | | |
| | | | 110 | 4.231 | 70 | 3.775 | 50 | 4.953 | | |
| | | | 150 | 4.535 | | | 90 | 5.066 | | |
| | | | 130 | 5.051 | | | | | | |
| | | | 30 | 1.037 | | | | | | |

Note CT Cure time, in seconds
 BS Bond strength, in MPa

Table 20 Bond Strength of U-F Adhesive Extended with Phosphorylated Sago at Several Temperatures

| Material grams | UF resin 6.1 | | Catalyst 0.43 | | Phosphorylated sago 3.38 | | Water 0.66 | | | |
|-------------------|-----------------|----|------------------|-------|-----------------------------|-------|---------------|-------|--------------|-------|
| Temp. °C. Code | 84 P85T | | 96 P95T | | 104 P105T | | 117 P115T | | 127 P125T | |
| | CT | BS | CT | BS | CT | BS | CT | BS | CT | BS |
| 100 | 2.269 | | 100 | 3.709 | 80 | 4.314 | 80 | 4.463 | 80 | 5.045 |
| 130 | 2.597 | | 120 | 4.525 | 110 | 5.589 | 110 | 4.997 | 100 | 5.013 |
| 160 | 2.668 | | 140 | 5.391 | 140 | 5.059 | 140 | 6.641 | 70 | 4.528 |
| 140 | 4.156 | | 160 | 4.989 | 120 | 6.175 | 120 | 4.997 | 50 | 4.470 |
| 120 | 3.341 | | 130 | 4.508 | 100 | 5.088 | 100 | 4.511 | 30 | 3.667 |
| 80 | 2.173 | | 110 | 3.532 | 70 | 3.699 | 90 | 4.515 | 20 | 2.599 |
| 60 | 1.368 | | 90 | 3.302 | 50 | 3.393 | 70 | 4.537 | 15 | 2.737 |
| 40 | 0.735 | | 70 | 1.956 | 30 | 1.985 | 50 | 4.488 | 40 | 4.490 |
| 20 | 0.044 | | 50 | 1.923 | 20 | 1.099 | 30 | 2.675 | 60 | 3.781 |
| 110 | 3.674 | | 30 | 1.833 | 60 | 2.394 | 20 | 2.078 | 55 | 4.425 |
| 90 | 2.372 | | 20 | 0.955 | 90 | 4.434 | 15 | 1.230 | 35 | 3.659 |
| 115 | 2.986 | | 60 | 2.460 | | | 40 | 3.995 | 45 | 4.477 |
| | | | 80 | 3.205 | | | 60 | 4.465 | | |
| | | | 40 | 1.842 | | | 35 | 2.838 | | |

Note: CT Cure time, in seconds
BS Bond strength, in MPa

Table 21 Bond Strength of U-F Adhesive Extended with
Wheat Flour at Several Temperatures, in MPa

| Material grams | UF resin 6.1 | | Catalyst 0.43 | | Wheat Flour 3.38 | | Water 2.6 | | | |
|-------------------|-----------------|----|------------------|-------|---------------------|-------|--------------|-------|--------------|-------|
| Temp. °C. Code | 84 P85T | | 96 P95T | | 104 P105T | | 117 P115T | | 127 P125T | |
| | CT | BS | CT | BS | CT | BS | CT | BS | CT | BS |
| 100 | 2.437 | | 100 | 2.671 | 80 | 3.521 | 80 | 3.808 | 80 | 5.648 |
| 130 | 2.597 | | 130 | 4.508 | 110 | 4.202 | 110 | 4.458 | 100 | 5.532 |
| 160 | 3.353 | | 160 | 5.032 | 140 | 4.465 | 130 | 5.177 | 70 | 5.046 |
| 190 | 3.703 | | 140 | 3.818 | 160 | 5.026 | 150 | 6.608 | 60 | 4.439 |
| 170 | 2.333 | | 120 | 3.030 | 120 | 4.458 | 120 | 5.099 | 50 | 4.355 |
| 140 | 2.671 | | 80 | 2.341 | 100 | 4.637 | 90 | 5.016 | 40 | 4.145 |
| 80 | 2.386 | | 60 | 2.104 | 80 | 3.608 | 60 | 3.768 | 30 | 3.626 |
| 60 | 1.185 | | 40 | 1.414 | 60 | 2.820 | 40 | 3.037 | 20 | 2.651 |
| 40 | 0.855 | | 20 | 0.477 | 40 | 2.087 | 20 | 1.706 | 15 | 1.570 |
| 20 | 0.020 | | 150 | 3.957 | 20 | 1.237 | 15 | 0.865 | 90 | 5.032 |
| 70 | 1.694 | | 125 | 2.805 | 90 | 4.541 | 115 | 4.345 | | |

Note: CT Cure time, in seconds
BS Bond strength, in mPa

Table 22 Bond Strength of U-F Adhesive Extended with Natural Sago Cured at 84 and 127 °C and Measured at Hot and Cool Temperature, in MPa

| 84 °C | | | | 127 °C | | | |
|-------|-------|-----|-------|--------|-------|-----|-------|
| Cool | | Hot | | Cool | | Hot | |
| CT | BS | CT | BS | CT | BS | CT | BS |
| 60 | 0.998 | 100 | 1.730 | 60 | 6.843 | 80 | 4.494 |
| 80 | 2.185 | 130 | 2.532 | 60 | 6.769 | 100 | 5.097 |
| 120 | 5.538 | 160 | 3.778 | 40 | 6.248 | 110 | 5.086 |
| 110 | 4.444 | 180 | 4.332 | 30 | 4.472 | 60 | 4.380 |
| 90 | 2.048 | 140 | 2.552 | 20 | 5.595 | 40 | 4.510 |
| 100 | 4.575 | 110 | 1.933 | 15 | 3.746 | 20 | 3.327 |
| 70 | 2.237 | 80 | 1.461 | 55 | 6.405 | 15 | 2.550 |
| 97 | 3.642 | 60 | 0.669 | 45 | 6.202 | 30 | 4.296 |
| 93 | 3.420 | 40 | 0.103 | 35 | 5.634 | 50 | 3.761 |
| 95 | 3.643 | 150 | 2.304 | 25 | 4.926 | 70 | 6.712 |
| 91 | 2.590 | 155 | 4.481 | 15 | 4.970 | 90 | 5.543 |
| | | 20 | 0.042 | 10 | 2.561 | 25 | 4.317 |
| | | | | 20 | 5.089 | 70 | 5.040 |
| | | | | 30 | 5.118 | | |
| | | | | 5 | 0.753 | | |
| | | | | 18 | 4.662 | | |
| | | | | 8 | 1.627 | | |
| | | | | 22 | 5.099 | | |

Note: CT Curing time, in seconds
 BS Bond strength, in MPa
 Cool bond is pulled at room temperature
 Hot bond is pulled at cure temperature

Table 23 Bond Strength of U-F Adhesive Extended with Phosphorylated Sago Cured at 84 and 127 °C and Measured at Hot and Cool Temperature, in MPa

| 84 °C | | | | 127 °C | | | |
|-------|-------|-----|-------|--------|-------|-----|-------|
| Cool | | Hot | | Cool | | Hot | |
| CT | BS | CT | BS | CT | BS | CT | BS |
| 40 | 2.129 | 100 | 2.269 | 30 | 4.937 | 80 | 5.045 |
| 80 | 4.475 | 130 | 2.597 | 40 | 7.255 | 100 | 5.013 |
| 100 | 4.889 | 160 | 2.668 | 20 | 4.505 | 70 | 4.528 |
| 120 | 5.041 | 140 | 4.156 | 10 | 2.795 | 50 | 4.470 |
| 140 | 5.536 | 120 | 3.341 | 15 | 4.900 | 30 | 3.667 |
| 70 | 3.622 | 80 | 2.173 | 35 | 4.561 | 20 | 2.599 |
| 65 | 3.296 | 60 | 1.368 | 25 | 5.063 | 15 | 2.737 |
| 30 | 2.128 | 40 | 0.735 | 35 | 5.638 | 40 | 4.490 |
| 55 | 4.385 | 20 | 0.044 | 20 | 4.779 | 60 | 3.781 |
| 50 | 2.153 | 110 | 3.674 | 40 | 6.117 | 55 | 4.425 |
| 60 | 2.627 | 90 | 2.372 | 23 | 4.563 | 35 | 3.659 |
| | | 115 | 2.986 | 27 | 4.516 | 45 | 4.477 |
| | | | | 14 | 4.334 | | |
| | | | | 12 | 3.246 | | |
| | | | | 32 | 4.940 | | |
| | | | | 117 | 7.907 | | |
| | | | | 3 | 0.541 | | |

Note: CT Curing time, in seconds

BS Bond strength, in MPa

Cool bond is pulled at room temperature

Hot bond is pulled at cure temperature

Table 24 Bond Strength of U-F Adhesive Extended with
Wheat Flour Cured at 84 and 127 °C and
Measured at Hot and Cool Temperature, in MPa

| 84 °C | | | | 127 °C | | | |
|-------|-------|-----|-------|--------|-------|-----|-------|
| Cool | | Hot | | Cool | | Hot | |
| CT | BS | CT | BS | CT | BS | CT | BS |
| 40 | 1.942 | 100 | 2.437 | 50 | 6.200 | 80 | 5.648 |
| 60 | 3.337 | 130 | 2.597 | 40 | 5.612 | 100 | 5.532 |
| 80 | 3.689 | 160 | 3.353 | 30 | 5.546 | 70 | 5.046 |
| 100 | 4.441 | 190 | 3.703 | 20 | 4.508 | 60 | 4.439 |
| 120 | 4.443 | 170 | 2.333 | 10 | 2.623 | 50 | 4.355 |
| 140 | 4.485 | 140 | 2.671 | 15 | 3.691 | 40 | 4.145 |
| 90 | 3.715 | 80 | 2.386 | 25 | 5.052 | 30 | 3.626 |
| 70 | 2.659 | 60 | 1.185 | 35 | 5.002 | 20 | 2.651 |
| 50 | 1.541 | 40 | 0.855 | 45 | 5.913 | 15 | 1.570 |
| 30 | 1.019 | 20 | 0.020 | 35 | 5.599 | 90 | 5.032 |
| 55 | 1.942 | 70 | 1.694 | 25 | 4.989 | | |
| 35 | 1.113 | | | 5 | 0.820 | | |
| 47 | 1.952 | | | | | | |

Note: CT Curing time, in seconds
BS Bond strength, in MPa
Cool bond is pulled at room temperature
Hot bond is pulled at cure temperature

Table 25 Bond Strength of U-F Adhesive Cured at 125° C
Measured at Hot and Cool Temperature, in MPa

| Cool | | Hot | |
|------|-------|-----|-------|
| CT | BS | CT | BS |
| 60 | 5.007 | 20 | 2.790 |
| 80 | 6.209 | 40 | 4.880 |
| 120 | 6.144 | 60 | 5.535 |
| 100 | 7.933 | 80 | 5.479 |
| 90 | 5.011 | 70 | 4.505 |
| 70 | 5.607 | 50 | 3.822 |
| 50 | 5.496 | 30 | 3.072 |
| 30 | 4.979 | 15 | 1.424 |
| 20 | 3.529 | 10 | 0.620 |
| 40 | 5.127 | 17 | 2.047 |
| 85 | 5.666 | 27 | 3.472 |
| 25 | 4.983 | 25 | 3.011 |
| 20 | 4.640 | 35 | 2.999 |
| 65 | 6.791 | 45 | 4.393 |
| 55 | 5.707 | 37 | 4.508 |
| | | 55 | 5.008 |
| | | 50 | 3.922 |
| | | 35 | 3.282 |
| | | 65 | 4.520 |

Note: CT Cure time, in seconds
BS Bond Strength, in MPa