FIG. 1(a)—Face checking of untreated resin-bonded Douglas-fir plywood after six months of exposure to the weather.

Wood- and Paper-Base Plastics

By ALFRED J. STAMM[1]

Forest Products Laboratory[2]

THE Forest Products Laboratory has been actively engaged during the last five years in the development of plastics from wood. Hitherto wood has been used almost exclusively in the forms of wood flour and wood pulp fillers for molding compounds and pulp preforms and in the form of paper for paper-base nonstructural laminates. Due to the efforts of the Forest Products Laboratory, wood is now finding use as the continuous phase in resin-treated, uncompressed wood ("impreg") and resin-treated, compressed wood ("compreg"). The plastic properties of the lignin constituent of wood are beginning to be utilized. Wood is being partially hydrolyzed to free the lignin and remove undesirable hemicelluloses in the making of molding powders which require smaller quantities of critical phenolic resin than do older commercial phenolic molding powders, its resin economy being due to the fact that the lignin serves as a plastic constituent (hydroxylin). Improved resin-impregnated paper-base laminates are being made that have practically twice the strength of earlier commercial paper-base laminates ("papreg").

All these newly-developed materials have strength properties comparable to or well above those of former plastics, and some of them are sufficiently strong for various military and peacetime structural uses.

IMPREG

A number of desirable properties can be imparted to wood by the forming of synthetic resins throughout the structure from resin-forming constituents of low molecular weight that have an affinity for wood. Although the hardness and compressive strength properties of wood can be improved by mechanically depositing any solid material within the structure, permanent dimensional stability and related properties have been successfully imarted to the wood only with a few specific resinoids under specific treating conditions.

Putting preformed resins into the structure merely blocks the entrance and exit of water, and hence, merely changes the rate of swelling and shrinking. Starting with a raw polar resin-forming mix, in a water solution, on the other hand, and allowing this intimately to penetrate the cell wall structure and bond to the active groups of wood, followed by evaporating off the water solvent and then heating to set the resin permanently, reduces the hygroscopicity of wood.

The most effective treating agent thus far found is a phenol-formaldehyde, water-soluble resinoid that is not advanced beyond the phenol-alcohol stage. Resorcinol can be substituted for the phenol or furfural for the formaldehyde without loss of effectiveness. All urea-formaldehyde resinoids tried have proved to be too highly prepolymerized to penetrate the structure adequately, with the exception of dimethylol urea. Even this material when polymerized within the structure reduced the swelling and shrinking on an equilibrium basis to only 60 per cent of normal, in contrast to reductions to 30 per cent of normal effected by phenol-formaldehyde resin. None of the thermoplastic resins or thermoplastic resin-forming systems thus far tried have effectively reduced the swelling and shrinking of wood, presumably because none of them have the desired affinity for the wood.

Difficulty has been encountered in properly distributing resin-forming chemicals throughout the structure of massive pieces of wood. The treatment appears practical only for veneer. The value of antishrink treatments of veneer which is normally built up into plywood might be questioned on the basis that, in cross-banded plywood, the fiber direction of one ply restrains the across-the-

[1]Presented before the Society of the Plastics Industry, New York, November 8 and 9, 1943.

[2]Maintained at Madison, Wis., in cooperation with the University of Wisconsin.

fiber dimension changes of the adjacent ply, thus mechanically reducing such changes. Swelling and shrinking cannot, however, be prevented mechanically. The mechanical restraint merely changes the direction of swelling and shrinking. If the wood is prevented from swelling normally in the sheet directions, it will swell in the thickness direction or internally into the fiber cavities. When normal plywood takes up and then loses moisture, the plies are continually working and, as a result of the unevenly developed stresses, face checking is more serious than in solid boards. Resin treatment, which reduces the swelling and shrinking to about 30 per cent of normal, reduces the stresses to such an extent that checking is practically eliminated. Fig. No. 1 shows two panels of Douglas-fir plywood that were exposed to the weather for six months without any surface finish. The contrast between the resin-treated and untreated surfaces is striking. The face checking of fancy crotch veneer for use in furniture and paneling can be similarly reduced by treatment with a phenolic resin.

The treatment of wood with stabilizing resins also imparts appreciable decay and termite resistance. Fig. No. 2 shows a three-ply piece of Douglas-fir plywood with treated faces and an untreated core. This specimen was immersed for six months to a depth of half its length in a field where decay and termite action on wood are severe. There was little sign of decay, but plenty of termite action. The termites in a frontal attack found the resin-treated faces not to their liking; so they tried a flank attack and, as a result, practically cleaned out the untreated core. Similar specimens that were edge-coated with resin and those in which all the plies were treated were still sound after a two-year exposure. The marked reduction in decay and termite action, it appears, is due rather to the fact that the treated wood will not take up enough water within the cell-wall structure to support decay than to the toxicity of the resin.

The treatment of wood with stabilizing resins increases its electrical resistance as a result of the reduced hygroscopicity. Dry wood is an excellent electrical insulator, but it loses its resistance properties rapidly with an increase in moisture content. At 30 per cent relative humidity the electrical resistance of the treated wood is about 10 times that of untreated wood, while at 90 per cent relative humidity it is about 1000 times as resistant.

Resin treatment also increases the acid resistance of wood, but it does not improve the alkali resistance.

The treatment of wood with 20 per cent of its weight of resin may increase the compressive strength and hardness by as much as 50 per cent. Most of the other strength properties are affected but slightly.

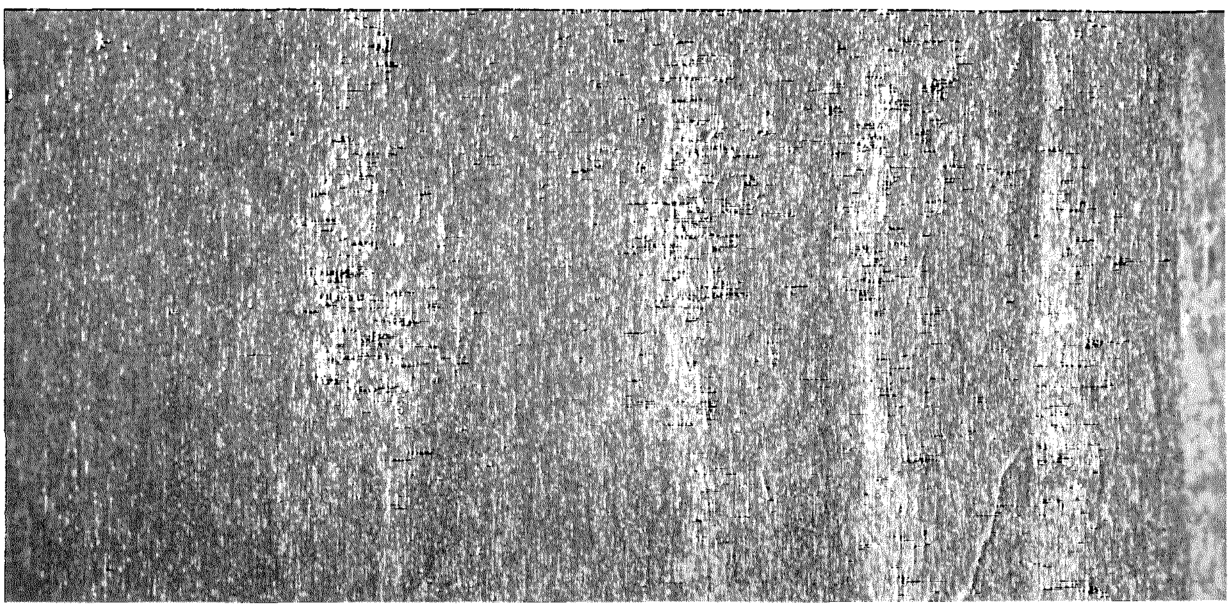
Impact strength is the one property that is adversely affected. As the resin content of wood is increased, it becomes more brittle. Likewise, more uniform distribution of the resin also increases brittleness. Unfortunately, the best treatment from the standpoint of stabilization is the poorest from the standpoint of brittleness. Normal birch has an Izod impact value of nine to 10 foot-pounds per inch of notch, but when treated with a stabilizing resin this value drops to only two to three foot-pounds per inch of notch.

COMPREG

Compreg is resin-treated wood that is compressed while the resin is formed within its structure. Although a number of different resins have been tried in making this material, none has proved as successful as phenol-formaldehyde. There are two types of compreg: 1. The older form, developed in Europe, which is treated with a spirit-soluble phenolic resin prepolymerized to the stage that it does not tend to penetrate the cell-wall structure and bond to the polar groups of the wood and, as a result, does not stabilize wood appreciably. 2. The form developed by the Forest Products Laboratory, which is treated with a water-soluble, phenol-formaldehyde resinoid, as in the case of impreg, so as to form the resin throughout the cell-wall structure of the wood and bond it to the active polar groups of the wood. The latter form of compreg is much more stable than the former but tends to be more brittle; like impreg, it has good decay and termite resistance and good electrical resistance.

Forest Products Laboratory compreg can be compressed to virtually the ultimate compression (specific gravity of 1.3 to 1.4) under a pressure of 1,000 pounds per square inch, using practically any species of wood. The unstable form of compreg, on the other hand, requires pressures of 2,500 to 3,000 pounds per square inch to compress the wood to the same degree. There is a still greater difference in the pressures required to compress the wood of the stable and unstable forms to intermediate degrees of compression. Practically all

FIG. 1(b)—Surface plies treated with 30 per cent by weight of synthetic resin (on the basis of the dry weight of the untreated wood).



the softwoods (coniferous woods) and the softer hardwoods (deciduous woods) such as cottonwood, basswood, and aspen, can, when treated with a stabilizing resin, be compressed to about one-half their original thickness under pressures as low as 250 pounds per square inch. This makes possible the compression of compreg faces and their simultaneous assembly with an untreated or treated and precured core with but slight compression of the core. This type of material, which shows great promise for postwar uses, cannot be made in one operation when the plies are treated with an appreciably polymerized resin, as there is little differential compressibility between such treated plies and the untreated plies.

A high degree of polish can be imparted to any cut surface of the Forest Products Laboratory form of compreg by merely sanding and buffing the surface. The potential finish exists throughout the structure. All that is necessary to bring it out is to smooth the surface. This easy way of restoring the finish would be an advantageous property of compreg or compreg-faced furniture. The natural finish is highly resistant to such organic solvents as alcohol and acetone, which destroy most applied finishes.

The water absorption of Forest Products Laboratory compreg is both small and slow. The water absorbed by a three-inch by one-inch by ⅜-inch specimen (one inch in the fiber direction) after immersion for 24 hours is less than one per cent. The unstabilized or less stabilized forms may absorb six per cent or more of water under the same conditions.

Forest Products Laboratory compreg will swell only four to seven per cent in thickness upon prolonged immersion in water at room temperature. Blocks less than an inch long in the fiber direction—the direction in which moisture absorption is greatest—will hardly come to swelling equilibrium when soaked in water at room temperature for a year. When dried to the original moisture content this compreg will practically regain its original dimensions, indicating that the loss of compression is negligible.

The unstable form of compreg will not only swell about three times as much in thickness as the stable form, but will swell much more rapidly and also lose a large part of its compression. The more rapid swelling is presumably due to the fact that water is sucked into the structure as it recovers from compression, and as a result water is distributed throughout the structure much more rapidly than by diffusion alone. The combined swelling and recovery from compression of the older form of compreg one inch long in the fiber direction may be as much as 20 to 60 per cent in several weeks. One-half to two-thirds of this dimensional change may be due to recovery from compression.

Most of the mechanical properties of the two forms of compreg are quite similar and, in general, vary almost in direct proportion to the compression. When wood is compressed to one-third of its original volume, its tensile strength, modulus of rupture in bending, and modulus of elasticity are about trebled, irrespective of whether the wood contains resin. Resin treatment prior to compression improves only the compressive properties and the shear strength in a plane at right angles to the direction of compression. Neither of these improvements, it is believed, is sufficient to warrant resin treatment unless it is accompanied by other improvements such as that of dimensional stability. Table No. I gives the normal strength properties of compreg in round figures.

The impact strength of compreg, like that of impreg, decreases with an increase in the resin content and the intimacy of distribution of the resin, although impact strength, unlike the other properties, will vary to a certain degree with variations in the processing conditions. Overheating during drying after resin treatment or in the pressing process tends to make the product more brittle. Under carefully controlled conditions, the stable form of compreg can be made from birch with

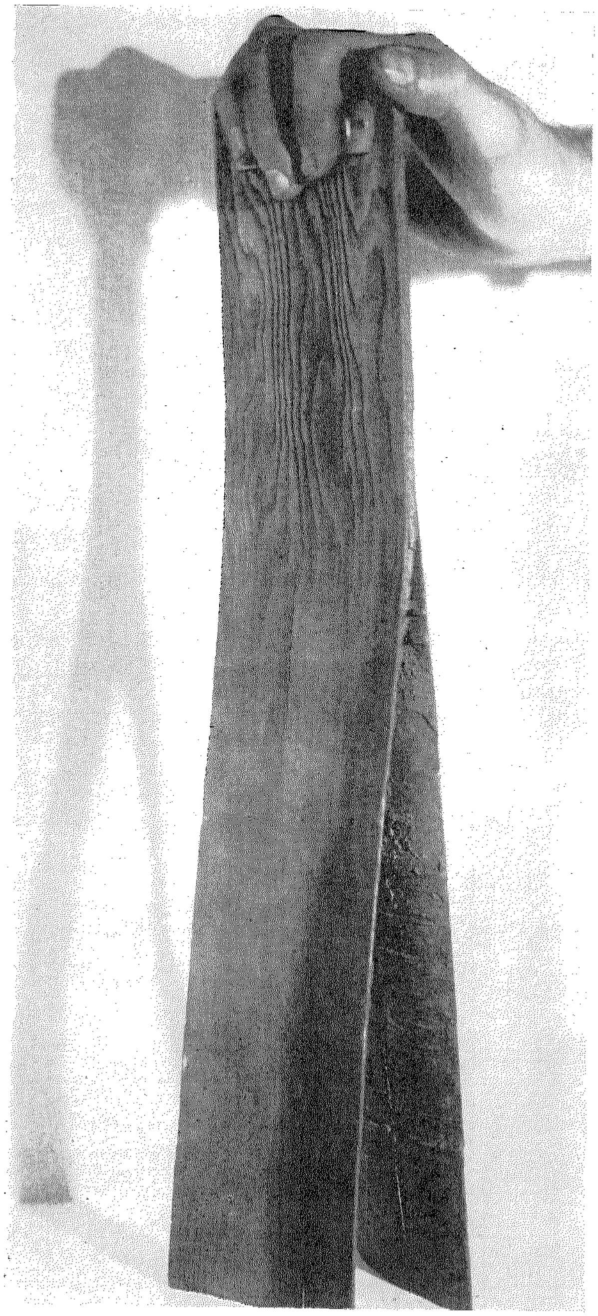


FIG. 2 — Action of termites on three-ply resin-bonded Douglas-fir plywood with faces treated with 30 per cent by weight of synthetic resin (on the basis of the dry weight of the untreated wood) and an untreated core that was immersed to half its length in a termite-infested field. The core has been almost completely eaten out up to the ground line while the faces are perfectly sound.

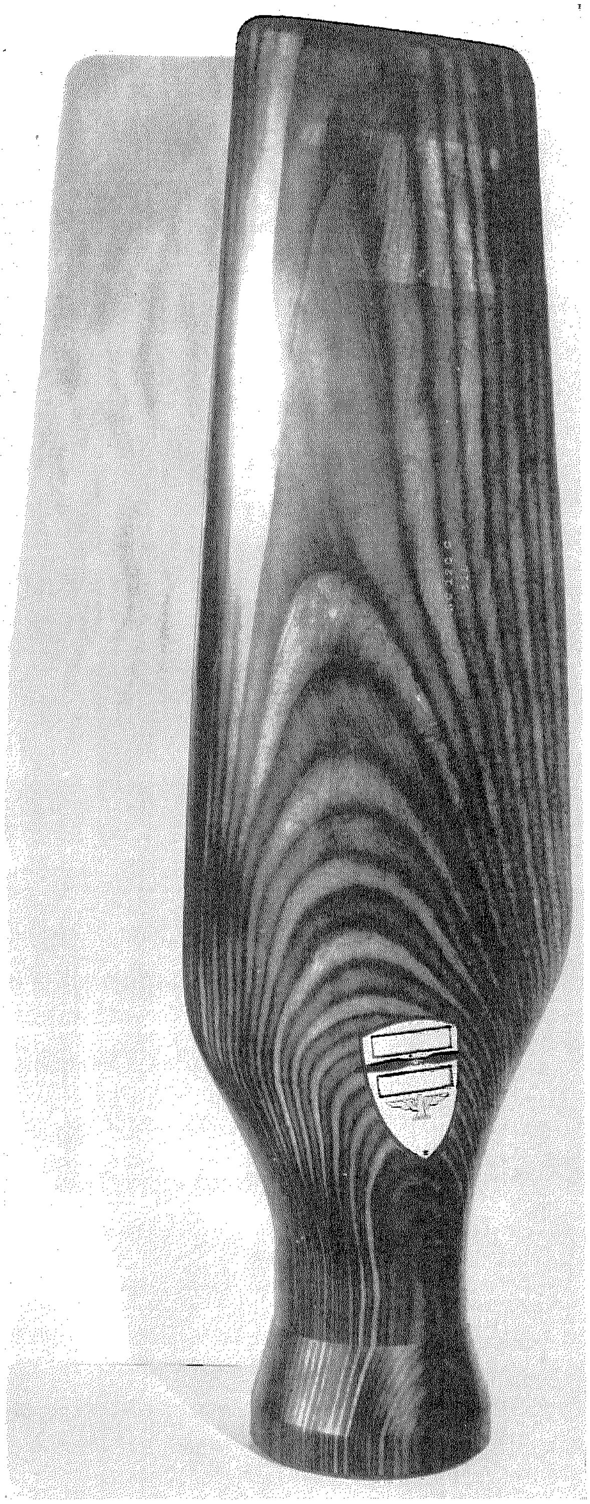


FIG. 3—Molded compreg propeller for testing airplane motors, now being commercially produced by a Michigan company using the process developed at the Forest Products Laboratory.

an Izod impact value of five to seven foot-pounds per inch of notch. The unstable compreg, on the other hand, will have an Izod value of six to nine foot-pounds per inch of notch.

TABLE I—NORMAL APPROXIMATE STRENGTH PROPERTIES OF PARALLEL-LAMINATED BIRCH COMPREG WITH A SPECIFIC GRAVITY OF 1.35[1]

Property	Value
	Lb. per sq. in.
Tension:	
Stress at proportional limit	22,000
Maximum strength	32,000
Modulus of elasticity	3,500,000
Flexure:	
Stress at proportional limit	21,000
Modulus of rupture	36,000
Modulus of elasticity	3,500,000
Compression parallel to grain:	
Stress at proportional limit	16,000
Maximum strength	24,000
Modulus of elasticity	3,500,000
Johnson double shear, parallel to grain and perpendicular to laminations	7,000
	Ft.-lb. per in.
Izod impact:[2]	
Face-notched	3 to 9
Edge-notched	2 to 7

[1]The properties given, with the exception of impact strength, are about the same for both stabilized and unstabilized compreg and do not vary appreciably between species.

[2]Three to seven foot-pounds per inch of notch for stabilized compreg (face-notched). Six to nine foot-pounds per inch of notch for unstabilized compreg (face-notched).

An important feature of compreg is that it can be made from a great variety of woods, including such normally inferior species as cottonwood, and obtain a product with properties which approach the optimum values. The only species to be avoided are the naturally resinous woods, such as southern pine, and those that are extremely difficult to treat, such as oak.

Compreg can be machined easily with metalworking tools but not with woodworking tools. Because of this, it is desirable to rough out the shape of objects prior to compression, using woodworking tools, and then compress them to the final shape in some form of mold. A technique for doing this has been developed at the Forest Products Laboratory. Treated, uncompressed plies are glued up into a blank of the correct size with a phenolic glue under conditions such that the treating resin is not cured and the bonding resin is but slightly cured. The shearing strength of such a block is not great but it is sufficiently strong so that it can be carved or turned in such a manner that the final dimensions are obtained in one plane, but the thickness at right angles to this plane is 1.5 to three times the final dimensions. The carved blank is then pressed in a split mold in the thickness direction. A Michigan company is using this method to mold propellers for the ground testing of airplane motors (Fig. No. 3) and airplane aerial masts. An airplane tail wheel has been successfully molded in this way so as to pass all static tests requirements (Fig. No. 4). The technique could be readily applied in the molding of pulley and gear wheels by stamping out the correct sections in the plane of the wheels from the individual plies and rotating these with respect to each other in the assembly as desired. Although wood is not moldable in the sense that a molding powder is, it is surprisingly subject to molding under proper conditions.

A recently-developed process of which nothing can at present be divulged makes possible the production of a highly stable form of compressed wood without the use of any impregnating resin.

HYDROXYLIN

Lignin is nature's plastic which cements the cellulose fibers of wood together. A mild hydrolysis treatment breaks the cellulose-lignin bond of wood, freeing the lignin so that it can be used to rebond the cellulose fibers together. Wood waste, preferably hardwood sawdust or mill waste, can be hydrolyzed by several different methods. The procedure which has received the greatest attention at the Forest Products Laboratory is a hydrolysis with dilute sulfuric acid in a rotary digester at a steam pressure of 135 to 200 pounds per square inch for 10 to 30 minutes. Besides breaking the cellulose-lignin bond, this hydrolysis treatment converts the hemicelluloses to sugars. These sugars, together with the acid, are washed out of the hydrolyzed wood and may be fermented to grain alcohol, thus giving a valuable by-product. The residue constitutes 50 to 60 per cent of the weight of the original wood. As a result of the removal of part of the cellulose, the lignin content is increased to 35 to 40 per cent.

After drying, the hydrolyzed wood is quite brash and can be readily ground to a powder, preferably of 40 to 100 mesh. Although the lignin in hydrolyzed wood can be made to flow sufficiently for the molding of some simple objects by merely adding small amounts of water and pressing at 375 degrees F., the flow is not adequate to give a product that is sufficiently coherent to stand long water immersion. Very similar results were obtained when nonresinous plasticizers for lignin were used in place of water, even though they did reduce the molding temperature. It was hence found necessary to use auxiliary plastics or plastic-forming constituents, together with a plasticizer for lignin, when the added plastic material did not also serve as such. The most suitable material found in the earlier work that served both functions is a mixture of eight per cent aniline and eight per cent furfural, together with 84 per cent of hydrolyzed wood and a small amount of mold lubricant such as zinc stearate. Molded products with good mold definition, water resistance, acid resistance, and electrical and mechanical properties can be obtained by pressing at 300 degrees F. for three minutes (in the case of small objects) at 3,000 to 4,000 pounds per square inch. Because the product is semithermoplastic, it must be cooled somewhat in the mold.

The flow of this molding powder is not so great as that of the general purpose commercial molding powders. This, together with the fact that the product cannot be drawn hot from the press, led to further research on the plasticizing of hydrolyzed wood. The best flow properties so far obtained have been with a molding powder containing 25 per cent of phenolic resin and 75 per cent of hydrolyzed wood. With this combination, the flow properties and the properties of the product are comparable with those of general-purpose molding compounds containing 50 per cent of phenolic resin and 50 per cent of wood flour. The fact that only half as much phenolic resin is required with the hydrolyzed wood as with the wood flour indicates that the lignin of the hydrolyzed wood imparts plastic properties to the product.

The hydrolyzed wood-phenolic resin molding powders give molded products with flexural strengths ranging from 8000 to 13,000 pounds per square inch, water absorptions of only 0.2 to 0.3 per cent after 48 hours' immersion in water, and extremely high acid resistance. It appears to be possible to mold this material into thicker flawless sections than can be made of general-purpose commercial molding powders. Because of these attributes, the material is now being tested in the molding of sizeable objects of industrial importance.

FIG. 4—Left to right, half of an airplane tail wheel molded of compreg; a compreg specimen varying in specific gravity from end to end (1.3 to 0.6); a model airplane propeller molded of compreg; a cut panel of birch compreg sanded and buffed to show that the finish exists throughout the structure.

If chips rather than sawdust are used as the raw material and the hydrolyzed product is abraded to a fiber rather than ground to a powder, it can be formed into a sheet on the paper machine. These sheets, with only a small amount of phenolic resin, can be compressed together into thick panels. The panels have considerably higher flexural strengths than panels made from the molding powder because of the reinforcing action of the much longer cellulose fiber.

PAPREG

Paper laminates treated with phenolic resins have been made for years. They have been used chiefly for electrical insulating panels and for other nonstructural uses which do not require exceptional mechanical properties. The manufacturers, in developing these materials, have approached the problem primarily from the resin standpoint. It was hence felt at the Forest Products Laboratory that further development of paper-base laminates, from the standpoint of finding the most suitable paper for the purpose, was a promising field of research. This proved to be the case. Within six months after the research was started, a paper-base laminate was developed that possessed several properties double those of the former laminates.

(*Continued on Page* 19)

remarkable differences are shown, for instance, by lettuce. Most varieties start to bolt soon after they are transferred to 80-degree night temperatures. In a 70-degree night temperature the plants also do not head properly, but very fine heads are obtained with lettuce kept during night at 45 and 55 degrees. The optimal day temperature for lettuce lies around 65 degrees. Even in tropical orchids the night temperature must be kept well below the day temperature, and most remarkable of all, the optimal temperatures for such orchids very closely approach the mean temperatures prevailing during day and during night in their natural surroundings.

METEOROLOGICAL DATA REQUIRED

If we ask now what these results mean for agriculture in general, there are several answers. In the first place, we gained a better insight into the conditions which determine growth and fruitfulness, so that we can give more precise directions for growing of tomatoes. These directions should include day and more particularly night temperatures. This leads to the second general conclusion: meteorological data, as usually presented to indicate the climate of a region, prominently display the *mean* temperature. This means very little from a tomato's viewpoint. The experiments described above indicate that the most important meteorological value for tomato growing would be the mean night temperature for each month. Extension of this line of research probably would lead to the most practical way of presenting meteorological data for agricultural use in general. In the third place, it becomes highly advisable to select better varieties of agricultural crops not in field trials, but in the synthetic climate of air-conditioned greenhouses. This conclusion is based on the observation that from year to year and from season to season tomato plants perform uniformly in air-conditioned greenhouses. Various synthetic climates, exemplifying main growing centers, could be created, and the best performers could be selected for each climate. Nowadays selection is made under natural conditions, which may mean a warm summer or a cool summer. The plants doing best one year in a warm summer probably will not do so well a subsequent year in a cool summer, so that selection under natural conditions becomes highly complex. By creating special Los Angeles area hot and cool weather varieties, and by utilizing future long-range weather forecasting, many of the hazards of agriculture could be eliminated, through use of the proper variety as indicated by the long-range weather forecast in spring. This would bring agriculture another step from the realm of art into the folds of engineering.

Wood- and Paper-Base Plastics

(*Continued from Page 9*)

Table No. II gives the readily obtainable properties of parallel-laminated papreg (machine direction of sheets all in same direction). Cross-banded papreg will have properties about two-thirds to three-fourths those of Table No. II.

Papreg has strength properties adequate for a large number of semistructural uses and some structural uses. As a structural material, its brittleness seems to be its most serious handicap. Compared to ordinary plastics, it has quite good Izod values, but it is definitely inferior in this respect to fabric and glass fabric laminates. It is, however, superior to fabric laminates in practically all other strength properties.

TABLE II—APPROXIMATE PROPERTIES OF PARALLEL-LAMINATED PAPREG

Property	Value
Specific gravity	1.38
	Lb. per sq. in.
Tension:	
Maximum strength	36,000
Modulus of elasticity	3,000,000
Flexure:	
Modulus of rupture	30,000
Modulus of elasticity	3,000,000
Compression:	
Parallel to grain	17,000
Flatwise perpendicular to grain	40,000
Edgewise perpendicular to grain	15,000
Johnson double shear, parallel to grain, perpendicular to laminations	13,000
	Ft.-Lb. per in.
Izod impact:	
Face-notched	5.0
Edge-notched	0.8
Hardness (Rockwell)	M 100
Water absorption (24 hours)	6 per cent

Because of its low elongation, papreg is not as easily molded to double curvatures as are fabric laminates. It has been successfully used, however, in molding of quite intricate objects with but a limited amount of goring and tailoring.

Work is now underway on incorporating other resins, both natural and synthetic, in paper-base plastics primarily from the standpoint of cheapening the product and also with the objective of building up the toughness without too great a sacrifice in water resistance and other mechanical properties. Details on this phase of the work cannot be given at present.

CONCLUSIONS

It is obvious from this array of products that wood is making an important place for itself in the plastics field. Although wood and its constituents serve mostly as the structural or filler part of these plastics, wood and wood products show promise of invading the resin field. Lignin and Vinsol (a rosin-purification residue) show promise as resin diluents. It is also of interest that phenols, furfural, and other resin-forming constituents are obtainable from wood by destructive distillation and hydrogenation processes. It does not require great imagination to visualize a self-contained wood industry that uses wood almost exclusively in the manufacture of wood plastics.

SOCIETY HONORS DR. MICHAL

The American Mathematical Society, at its 50th Annual Meeting in Chicago last week, elected Aristotle D. Michal a vice-president. The society is a national organization devoted in peacetime to mathematical research and during wartime to mathematical problems connected with the design of military equipment. Dr. Michal, a member of the society for 20 years, has been at the Institute since 1928 as a professor of mathematics.