

◀Original▶ A Study on the Preparation of Wood-Plastic Combinations (IV)

The Physical and Chemical Properties of Wood-Plastic Combinations

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Abstract

Some physical and chemical properties of wood-plastic combinations(W.P.C.) made of domestic soft woods such as *pinus densiflora*, *pinus rigida* and *populus deltoides* were measured.

The rates of improvement in properties were roughly proportional to the contents of polymer or polymer mixtures in W.P.C.

For the W.P.C. obtained by means radiation curing and containing 80% of polymer or polymer mixture, the hardness and water absorptivity were improved 2.2 times and 4 times those of the original wood, respectively. The improvement of hardness was especially remarkable in the W.P.C. made of *pinus densiflora* and polystyrene(120%) to show 7 times increased hardness.

For the W.P.C. obtained by means of thermal curing and containing 80% of polymer or polymer mixture, the hardness and water absorptivity were improved 2.4 times and 3.4 times those of the non-treated woods, respectively.

These data indicate that the properties of W.P.C. prepared by means of radiation curing are not much different from those of W.P.C. prepared by means of thermal curing.

Both of acid resistancy and alkali resistancy of the W.P.C. were also improved remarkably in comparison with the non-treated wood.

요 약

육송, 미송, 포플라등 국산 연질목재를 사용하여 만든 목재-플라스틱 복합체(W.P.C.)의 물리적 화학적 성질을 측정하였다.

성질 향상의 정도는 복합체에 함유된 중합물량에 대략 비례하였다. 중합물 함량이 나무무게에 대하여 80%되게 방사선 중합법으로 제조한 복합체에 있어서의 경도(硬度) 및 흡수도(吸水度)는 원래의 나무의 그것에 비하여 2.2배 및 4 로 각각 향상되었다. 경도의 향상은 육송에 폴리스타이렌을 120% 함유하게 만든 복합체에서 특히 현저하여 7배의 향상을 보였다.

중합물함량이 나무무게에 대하여 80%이하 열중합법으로 제조한 복합체에 있어서의 정도 및 흡수도는 나무만일때에 비하여 2.4배 및 3.4배로 각각 향상되었다.

이와같은 결과로 보아 방사선중합법으로 제조한 복합체와 열중합법으로 제조한 복합체에 있어서의 성질에는 별 차이가 없음을 알수 있었다.

그 밖에 내산성, 내알칼리성도 나무만일때에 비해 크게 향상 되었다.

1. Introduction

The generally known methods for preparing wood-plastic combinations(W. P. C.) are radiation curing¹⁻⁹⁾ and thermal curing^{10, 11)}.

The results of the studies on the preparations of W.P.C. using domestic soft wood have already been reported^{12, 13)}

In this paper the authors have investigated on the properties of W.P.C. prepared by both methods to evaluate and to compare the rates of improvement in hardness, water absorptivity, acid resistancy and alkali resistancy, in view of selecting suitable monomer, of determining the amounts of impregnation and of choosing the most suitable domestic soft wood. The up to date data concerning the properties of W.P.C. made of domestic soft wood are hardly available.

2. Experimental

(1) Materials and Equipment

a) Thermally cured W.P.C. samples; 1×1×10cm sized W.P.C. prepared by the method previously reported¹³⁾.

b) Radiation-cured W.P.C. samples; 1×1×10cm sized W.P.C. prepared by the method previously reported¹²⁾

c) Testing Instrument; Universal testing machine, Instron Model TML, maximum applicable load; 100kg.

(2) Procedure

a) *Measurement of Hardness*; the hardnesses were measured in normal direction to the annual ring of the wood stock and were expressed in B.H.N.(Brinell hardness number) according to the following equation;

$$B. H. N. = L/\pi \cdot D \cdot h$$

where D: dia.(mm) of the ball,

L: applied load(kg.),

h: depth(mm.) of the formed indentation

The conditions of the measurement of hardness were as followings;

applied load: 40kg.,

dia. of the ball: 7.925mm.,

cross head speed: 0.05cm/min.,

chart speed: 0.5cm/min.

b) *Measurements of Water-Absorptivity*

Various pre-weighed W.P.C. samples were immersed into 20°C water contained in thermostat. After 24 hrs, the samples were taken out and weighed again. The differences in weight were expressed as water-absorptivity.

c) *Measurements of Acid-Resistancy*

Various W.P.C. samples of known weight were immersed into 5% hydrochloric acid solution settled in thermostat. The temperature of the thermostat was adjusted to 20±0.5°C. After 8hrs., the temperature was raised to 80±0.5°C and maintained the samples immersed for 8 hrs.

After washing the samples with plenty of water, they were dried to constant weight and re-weighed. The hardness of the samples were also measured after the treatment.

d) *Measurements of Alkali Resistancy*

Various samples were treated in the similar way to that of c) with 5% sodium hydroxide solution. The samples were immersed in the solution for 10 hrs at 20±0.5°C and 1 hr at 100°C.

e) *Measurements of Dimensional Changes*

Various W.P.C. samples of known dimen-

sions were immersed into 30°C water for 48 hrs. and then dried to constant weight. The hardness in normal direction, in tangential direction and in lengthwise direction were measured, respectively.

3. Results and Discussion

(1) Improvement in Hardness

As Fig. 1 shows, the hardness is increased 6 times in pinus densiflora-(poly M. M. A. + poly V. Ac.(70-80%)) combination. It is reported by Mani¹⁴⁾ that the Fir-poly M. M. A. (135%) combination made by radiation curing method shows 7 times increased hardness. This value is nearly consistent with our data. Among the W.P.C. made of various wood and polystyrene, the pinus densifloraspolystyrene(100-110%) combination shows the most remarkably increased hardness.

On the other hand, the pinus rigida-polystyrene(100-110%) combination and poplus deltoides-polystyrene(100-110%) combination show only 2 times increased value, respectively(Fig. 2). Such a tendency is also found in the W.P.C. made of polystyrene and poly V. Ac; pinus densiflora-(polystyrene+poly V. Ac.(50%)) combination shows 2 times increased hardness but the the pinus rigida-(polystyrene +poly V. Ac.(50%)) combination and poplus deltoides-(polystyrene+poly V. Ac.(50%)) combination show 0.4 times and 1.8 times increased hardness, respectively(Fig. 3). As Fig. 1 to 3 show, the pinus densiflora-plastic combinations show highly increased hardness, and the hardness increase is especially significant when the wood is combined with poly M. M. A.(Fig. 1).

Thus, it may be said that the M. M. A. is suitable monomer to get hard W.P.C. The hardness increase in pinus rigida-plastic combination is inferior to the pinus densiflora-plastic combination regardless the kind of polymer.

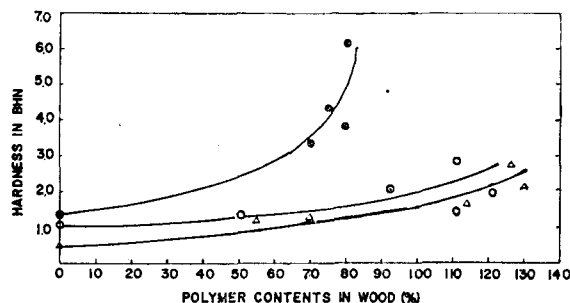


Fig. 1. Hardness vs. polymer contents in W. P.C. prepared by thermal curing method. polymer mixture; poly M. M. A. + poly V. A. -⊗-: pinus densiflora, -○-: pinus rigida, -△-: poplus deltoides

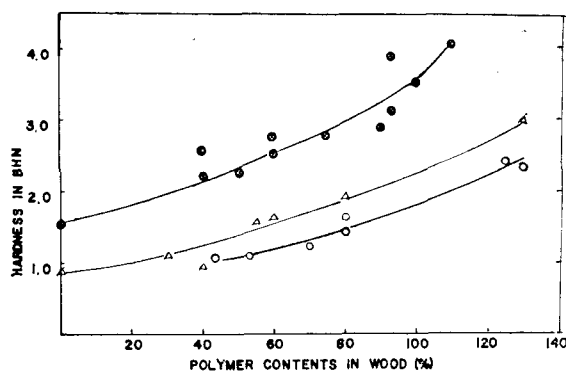


Fig. 2. Hardness vs. polymer contents in W. P.C. prepared by means of thermal curing method. polymer; polystyrene -⊗-: pinus densiflora, -○-: pinus rigida, -△-: poplus deltoides

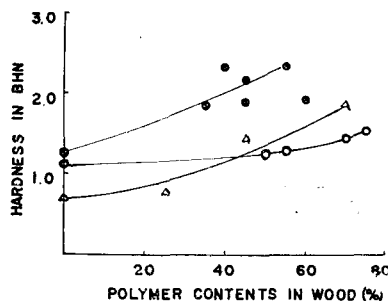


Fig. 3. Hardness vs. polymer contents in W. P.C. prepared by thermal curing method. polymer mixture; polystyrene + poly V. A. -⊗-: pinus densiflora, -○-: pinus rigida, -△-: poplus deltoides

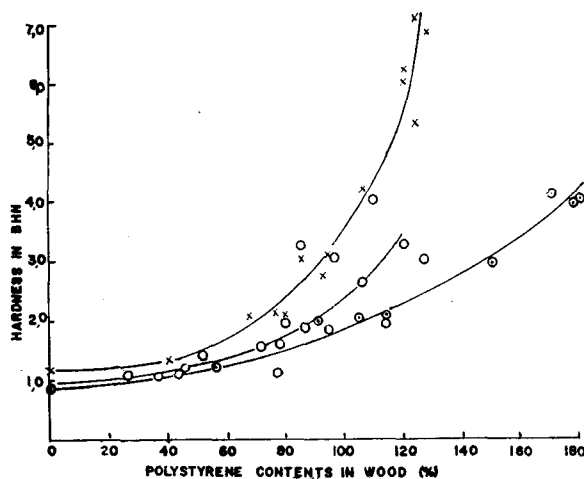


Fig. 4. Hardness of wood-polystyrene combinations prepared by radiation induced polymerization.
 -x-: pinus densiflora, -○-: white lauan,
 -⊙-: populus deltoides

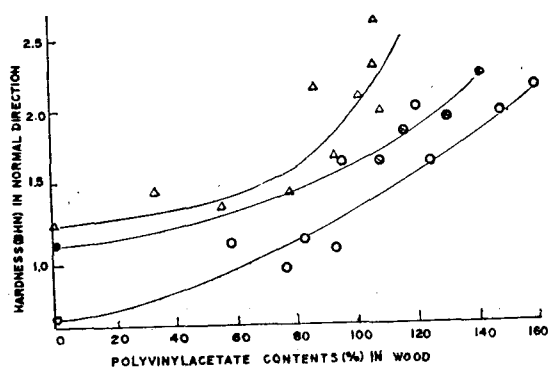


Fig. 5. Hardness of wood-poly V. A. combination prepared by radiation induced polymerization.
 -△-: pinus densiflora, -⊗-: populus rigida,
 -○-: populus deltoides

The hardness is increased 6 times that of the original wood in pinus densiflora polystyrene (120%) combination made by gamma irradiation method. However, the hardness increases are lower than this value in the W.P.C. made of other woods by the same method; *i. e.* white lauan-polystyrene(120%) combination and populus deltoides-polystyrene(120%) combination show hardness increases of 3 times and 2.5 times, respectively(Fig. 4). The hardness increase in normal direction to the

annual ring is also remarkable in the pinus densiflora-poly V. Ac (100%) combination made by gamma irradiation(Fig. 5). Therefore, it is confirmed that the tendency of hardness increase in the W.P.C. made by gamma irradiation is consistent with that in W.P.C. made by thermal curing; *i. e.* the hardness increase is especially remarkable in the pinus densiflora-plastic combinations made by both methods.

K.N. Rao¹⁵⁾ reported that the hardness improvements of Haldu-polystyrene(50%) combination and Teak-polystyrene(36%) combination are 1.7 times(from 3.2 B.H.N. to 5 B.H.N.) and 1.08 times(from 2.75 B.H.N. to 3 B.H.N.), respectively. According to these data, it may be said that the natural born hard woods are difficult to be impregnated with monomer, and even if impregnated, they are still difficult to be polymerized owing to the presence of a lot of resin in wood. Furthermore, even if they are polymerized in wood, the rate of hardness increase is quite small.

It is reported¹¹⁾ that the Santol-poly M. M. A. (46%) combination made by thermal curing method shows about 3.5 times increased hardness. This value is higher than the value obtained by us comparing the polymer contents in wood. According to the Mittinen's postulation¹¹⁾, the above data are not exactly comparable with ours since the specimen of the wood is different. They insisted that M. M. A. and mixtures of different unsaturated polyesters with styrene in about 1:1 ratio all give approximately similar strength properties if the wood materials are same.

(2) Improvement of Water Absorptivity

The original populus deltoides absorbs 60 to 70% of water based on the weight of wood when it is immersed into 20°C water for 24 hrs. However, the water absorptivity of populus

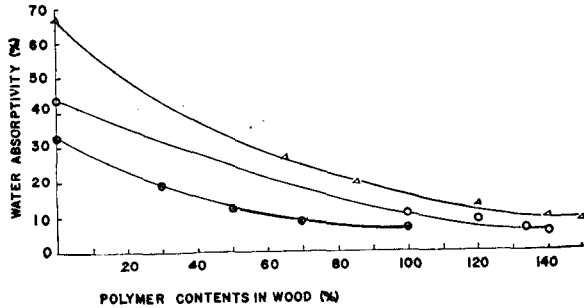


Fig. 6. Water absorptivity vs. polymer contents in W.P.C. made by thermal curing method.
 polymer mixture: poly M. M. A. + poly V. A.
 -⊗-: pinus densiflora-p.c., -○-: pinus rigida-p.c., -△-: populus deltoides-p.c.

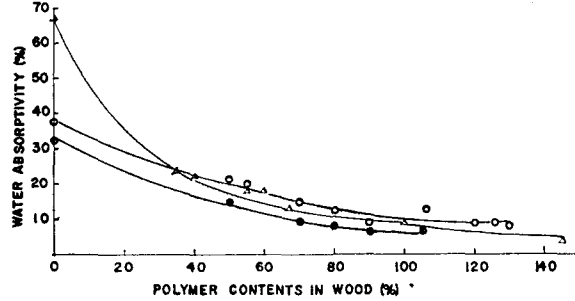


Fig. 9. Water absorptivity vs. polymer contents in W.P.C. prepared by thermal curing method.
 polymer; polystyrene
 -⊗-: pinus densiflora, -○-: pinus rigida,
 -△-: populus deltoides

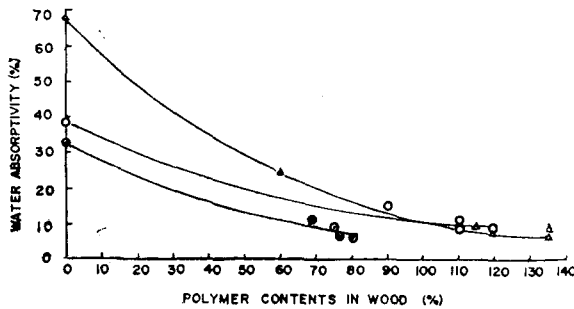


Fig. 7. Water absorptivity vs. polymer contents in W.P.C. made by thermal curing method.
 polymer mixture; polystyrene + poly V. A.
 -⊗-: pinus densiflora-p.c., -○-: pinus rigida-p.c., -△-: populus deltoides-p.c.

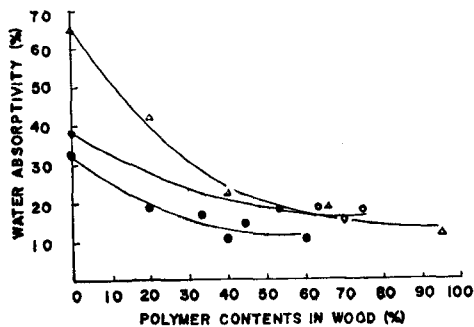


Fig. 8. Water absorptivity vs. polymer contents in W.P.C. made by thermal curing method.
 polymer mixture; poly M. M. A. + polystyrene,
 -⊗-: pinus densiflora-p.c., -○-: pinus rigida-p.c., -△-: populus deltoides-p.c.

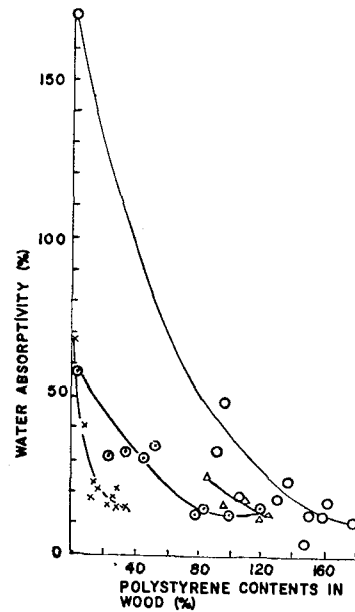


Fig. 10. Water absorptivity vs. polystyrene contents in wood-polystyrene combination prepared by radiation induced polymerization.
 -X-: yellow lauan, -⊗-: white lauan,
 -△-: pinus densiflora, -○-: populus deltoides

deltoides-(poly V. Ac+poly M. M. A. (130 %)) combination is only 8% under the same conditions of treatment(Fig. 6). The water absorptivity of the populus deltoides-(polystyrene+poly V. Ac.(100%)) combination is 10% (Fig. 7) and that of the populus deltoides-(poly M. M. A. +polystyrene(140%) combina-

Table 1. Acid resistancy of W.P.C. prepared by means of thermal curing method

Wood	Polymer	Polymer contents in wood (%)	Hardness (B. H. N.) of original wood	Weight decrease (%)	Hardness variation (%)
Pinus rigida	—	—	1.185	9.0	*↓ 31.15
Pinus rigida	Styrene	69.1	1.883	5.6	↓ 25.7
Pinus rigida	Styrene	132.8	2.107	2.2	↓ 23.7
Pinus rigida	MMA+styrene	66.2	2.093	7.7	↑ 21.7
Pinus rigida	MMA+styrene	118.4	3.370	2.9	↑ 19.3
Pinus rigida	VA+styrene	53.1	1.159	12.6	↓ 23.0
Pinus rigida	VA+styrene	71.1	1.347	11.6	↓ 3.8
Pinus densiflora	—	—	1.560	13.5	↓ 51.6
Pinus densiflora	Styrene	51.7	2.388	5.3	↓ 6.6
Pinus densiflora	Styrene	102.3	3.050	3.3	↑ 28.5
Pinus densiflora	MMA+styrene	49.3	2.810	4.9	↓ 9.2
Pinus densiflora	MMA+styrene	96.1	4.632	2.6	↓ 33.3
Pinus densiflora	VA+styrene	75.0	4.428	2.6	↓ 7.9
Pinus densiflora	VA+styrene	95.0	4.632	0.7	↑ 2.1
Poplus deltoides	—	—	0.950	16.1	↓ ∞
Poplus deltoides	Styrene	40.3	1.088	6.6	↓ 57.7
Poplus deltoides	Styrene	155.1	4.503	1.6	↓ 6.1
Poplus deltoides	MMA+VA	55.5	1.607	5.4	↓ 66.8
Poplus deltoides	MMA+VA	133.8	2.374	3.9	↓ 32.3
Poplus deltoides	VA+styrene	23.4	2.173	12.9	↓ 80.4
Poplus deltoides	VA+styrene	66.0	1.734	10.2	↓ 39.0

*↓ decrease ↑ increase

tion is also 10%(Fig. 8). About 90% of the original water absorptivity is decreased in the poplus deltoides-polystyrene(145%) combination showing the water absorptivity of only 4% under the same conditions of treatment (Fig. 9). Significant improvement in water absorptivity was also found by Mani¹⁴⁾; *i. e.*, the Fir-poly M. M. A.(135%) combination made by radiation curing method shows 11 times improved water absorptivity when the W.P.C. is treated in 25°C water for 24 hrs. The degree of water absorptivity decrease is similar between pinus rigida-plastic combination and pinus densiflora-plastic combination to show 75-80% decrease regardless the kind of the contained polymer material.

As for as the rate of the decrease in water absorptivity is concerned, the poplus deltoides-plastic combination seems to be better than pinus densiflora plastic combination, however,

when the amount of water absorptivity is only taken into account, the former is worse than the latter. Actually, the water absorptivity of the original pinus densiflora is 30%, but that of the pinus densiflora-plastic(100%) combination is only 5-8% regardless the kind of the polymer material(Fig. 6, 8, 9). Consequently, the water absorptivity decreasing rate is in the order of poplus deltoides-plastic combination > pinus rigida-plastic combination > pinus densiflora-plastic combination.

The water absorptivity of W.P.C. prepared by means of gamma irradiation is also remarkably decreased; the poplus deltoides-plastic (160%) combination shows 11 times decrease when it is treated in 30°C water for 48 hrs (Fig. 10).

10 times of water absorptivity decrease was observed by Ramalingam²⁾ when pine-polystyrene combination prepared by means of gamma

irradiation with 20°C water for 1 month. This value nearly coincides with ours obtained by the W.P.C. immersion into 30°C water for 4 hrs. It is expected that the W.P.C. prepared by means of thermal curing will show a little bit more improved values in case of immersion into water of higher temperature and for longer period.

Of course, the Ramalingam's value is not strictly comparable with ours because the exact contents of polymer in W.P.C. is not known. The values of water absorptivity decrease in W.P.C. made by means of gamma irradiation are similar to that in W.P.C. made by means of thermal curing; 94% of water absorptivity decrease is observed in the populus deltoides-polystyrene(180%) combination made by gamma irradiation method, and 90% decrease is observed in the populus deltoidespolystyrene+poly M. M. A. (150%) combination made by thermal curing method(Fig. 8.10).

(3) Improvement of Acid Resistancy

The acid resistancy of the original wood is very poor. The weight of populus deltoides, especially, is decreased 16% of the original weight and is crushed when the treated sample was applied to the hardness tester(Table 1). However, in populus deltoides-polystyrene(155%) combination the weight loss and the hardness decrease were only 1.6% and 6%, respectively. In pinus rigida-(poly M. M. A. + polystyrene) combination, the weight is decreased but the hardness is rather increased. This phenomenon may be attributable to the surface corrosion of the wood cellulose and consequent exposure of the plastics. Further, the hardness of wood cellulose itself or contents of resin may also affect to the hardness variation in acid treatment.

The mean improvement values in weight change in pinus rigida-plastic(85%) combination, pinus densiflora-plastic (78%) combin-

ation and populus deltoides-plastic (80%) combination are 20%, 70% and 57%, respectively. Consequently, the order of improvement in weight change is pinus densiflora-plastic combination > populus deltoides-plastic combination > pinus rigida-plastic combination. When the polymer contents are same the order of improvement in hardness change will be populus deltoides-plastic combination > pinus densiflora-plastic combination > pinus rigida-plastic combination. When the rates of improvement in hardness and in weight change are simultaneously considered the pinus densiflora and the populus deltoides are superior to the pinus rigida. When we consider the kinds of monomer used the rate of improvement of weight change in the combinations of wood-polystyrene, wood-(poly M. M. A. + polystyrene (87%) and wood-(poly M. M. A. + poly V. Ac (64%)) are, respectively, 60%, 54% and 14%. Upon these data, we deduce the general trend of improvement; the styrene or M. M. A. + styrene is superior to the styrene+V. Ac. The rate of improvement of hardness change in wood-polystyrene(88%) combination and in wood-(poly M. M. A. + polystyrene (82%)) combination are 67% and 100%, respectively. As the superiority of M. M. A. in the improvement of hardness is confirmed in the measurement of hardness improvement in W.P.C., the M. M. A. is also superior to the others in the rate of improvement of hardness change in acid resistancy.

(4) Improvement of Alkali Resistancy

In the pinus densiflora-plastic combination and pinus rigida-plastic combination the hardnesses are increased but the weights are decreased slightly(Table 2). In populus deltoides-plastic combination the changes in weight and hardness are more extreme than that in other W.P.C. but still they are minute comparing those obtained in the acid resistancy

Table 2. Alkali resistancy of W.P.C. Prepared by means of thermal curing method

Wood	Polymer	Polymer contents in wood (%)	Hardness (BHN) of original wood	Weight decrease (%)	Hardness variation (%)
Pinus rigida	—	—	1.185	3.3	*↓ 21.1
Pinus rigida	Styrene	76.5	1.343	1.5	↑ 16.2
Pinus rigida	Styrene	106.5	2.135	0.7	↑ 42.1
Pinus rigida	MMA+VA	90.0	2.254	1.8	↑ 18.6
Pinus rigida	MMA+VA	121.0	2.096	1.3	↑ 18.0
Pinus rigida	MMA+styrene	66.3	0.976	5.7	↑ 51.1
Pinus rigida	MMA+styrene	119.6	1.645	0.7	↑ 1.2
Pinus densiflora	—	—	1.560	3.1	↑ 3.0
Pinus densiflora	Styrene	58.3	2.738	1.9	↑ 17.4
Pinus densiflora	Styrene	105.1	4.049	0.5	↑ 16.8
Pinus densiflora	MMA+VA	68.1	3.517	1.5	↑ 15.7
Pinus densiflora	MMA+VA	79.3	6.020	0.5	↑ 3.5
Pinus densiflora	MMA+styrene	30.6	1.876	6.5	↑ 11.3
Pinus densiflora	MMA+styrene	67.0	4.153	1.5	↑ 7.9
Poplus deltoides	—	—	0.950	2.7	↓ 25.2
Poplus deltoides	Styrene	34.5	1.249	2.1	↓ 42.5
Poplus deltoides	Styrene	102.1	1.820	4.1	↓ 0.8
Poplus deltoides	MMA+styrene	62.7	0.834	4.9	↓ 44.7
Poplus deltoides	MMA+styrene	131.4	3.050	2.6	↓ 16.9
Poplus deltoides	VA+styrene	43.8	1.461	14.6	↓ 50.0
Poplus deltoides	VA+styrene	91.6	1.390	4.2	↑ 21.7

*↓ decrease, ↑ increase

measurement.

Alkaline hydrolysis of polymer or resin in the W.P.C. seems to be occurred during the alkali treatment. There was white solid material covered on most of the surface of the W.P.C. pieces after alkali treatment. It seems that the solid material makes the weight and hardness decreases less.

For combinations of pinus rigida-plastic(97%), pinus densiflora-plastic(68%) and poplus deltoides-plastics(78%), the rates of improvement in weight change are 30%, 36% and 78%, respectively. Thus, the order of improvement in weight change is poplus deltoides-plastic combination > pinus densiflora-plastic combination > pinus rigida-plastic combination. For the same combinations of the above, the rates of improvement in hardness change are 120%, 400% and 15%, respectively. Thus, the order of improvement in hardness change is

pinus densiflora-plastic combination > pinus rigida-plastic combination > poplus deltoides-plastic combination.

For the combinations of wood-polystyrene (80%), wood-(poly M.M.A.+polystyrene(90%)), and wood-(polystyrene+poly V. Ac (70%)), the rates of improvement in hardness change are 160%, 94% and 0%, respectively. Consequently, styrene or M.M.A.+styrene is superior to styrene+V. Ac in alkali resistancy.

(5) Improvement of Dimensional Stability

As Table 3 shows, the poplus deltoides-polystyrene(136%) combination brings about the most improved dimensional stability. This fact coincides well with the strong water absorptivity of the poplus deltoides and the great decrease of water absorptivity in the poplus deltoides-plastic combinations.

These results seem to give some promise

Table 3. Dimensional change % for wood-polystyrene combinations.

Direction to the annual ring	W.P.C.	p. d. / p. d.-p. s. (106.9%)	p. del. / p. del.-p. s. (136%)	a. p. / a. p.-p. s. (10.4%)	w. l. / w. l.-p. s. (84%)	y. l. / y. l.-p. s. (25%)	r. l. / r. l.-p. s. (10.3%)
	Tangential		6.13/2.84	7.06/1.85	4.76/4.42	3.96/1.90	3.96/1.45
Normal		5.56/2.48	3.76/1.13	6.36/5.41	4.21/2.37	4.21/2.70	3.92/3.58
Lengthwise		1.43/0.19	1.44/0.41	0.48/0.12	0.96/0.23	0.99/0.15	0.48/0.16

p. d. : pinus densiflora, p. del. : populus deltoides, a. p. : apitong, w. l. : white lauan, y. l. : yellow lauan, r. l. : red lauan, p. s. : polystyrene

that the properties of W.P.C. could be much improved in the further tests such as tensile strength, impact strength, fatigue strength under bending stress, abrasion resistance and weather resistance *etc.* However, these tests are not carried out in this work.

4. Conclusions

(1) The rate of improvement of physical and chemical properties of W.P.C. is increased with polymer contents in W.P.C.

(2) The most suitable monomer and wood to upgrade the hardness in W.P.C. are M. M. A. and pinus densiflora, respectively.

(3) The hardness of W.P.C. is improved 2 times that of the original wood when woods contain in average 70% of polymer.

(4) The rate of improvement of water absorptivity in populus deltoides-plastic combination is the most remarkable. In the populus deltoides-plastic(70-80%) combination the rate of improvement of water absorptivity is 70% under the given conditions.

(5) The properties of W.P.C. made by gamma irradiation method are not much different from those of W.P.C. made by thermal curing method as far as the polymer contents are similar.

(6) In the acid resistancy, the order of improving weight change is pinus densiflora-plastic combination > populus deltoides-plastic combination > pinus rigida-plastic combination. The order of improving hardness change is populus deltoides-plastic combination > pinus den-

siflora-plastic combination > pinus rigida-plastic combination.

(7) In alkali resistancy, the order of improving weight change is populus deltoides-plastic combination > pinus densiflora-plastic combination > pinus rigida-plastic combination.

(8) The rate of improvement in dimensional stability under the given conditions is the most remarkable in populus deltoides-plastic combination.

(9) When the rates of improving physical and chemical properties of W.P.C. are taken into account, the pinus densiflora and the populus deltoides are superior to pinus rigida, and M.M.A. and styrene are more effective than V. Ac.

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