

◀Technical Report▶ **A Study on the Preparation
of Wood-Plastic Combination (I)**

General Properties and Radiation Durabilities of Woods

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Abstract

General physical properties and radiation durabilities of some domestic woods and imported lauan were measured and discussed as a preliminary study on the preparation of W.P.C.

The data obtained indicate that 1) the hardness of wood is generally proportional to the specific gravity and to the bending strength, 2) the hardness of wood measured in tangential direction to the annual ring increases along with the distance from the center of the annual ring to the peel, 3) the changes of specific gravity under 100 Mrad gamma irradiation are not remarkable, 4) the hardnesses of the soft woods are decreased more readily than those of the hard woods, but the decreases of hardnesses are not extreme upto 10 Mrad suggesting that the woods are sufficiently durable to gamma irradiation of below 10 Mrad in the preparation of W.P.C.

요 약

국산 목재와 수입라완의 물리적 성질과 방사선 내성을 측정하여 W, P, C 제조 예비 실험을 한 결과 다음과 같은 결과를 얻었다.

- 1) 목재의 경도는 비중과 휨강도 (bending strength)에 비례했다.
- 2) 연륜(年輪) 접선 방향의 경도는 목심(木心)으로부터의 거리에 비례했다.
- 3) 100 Mrad의 감마선 조사로는 목재의 비중에 큰 변화가 생기지 않았다.
- 4) 방사선 조사시 연질목재의 경도가 경질 목재의 그것보다 더욱 현저히 감소했다.

그러나 10 Mrad까지는 경도 감소가 심하지 않음으로 보아 W, P, C 제조에 있어 10 Mrad 이하의 조사에는 목재가 충분한 내성을 가짐을 알 수 있다.

1. Introduction

The preliminary investigation on the general properties and radiation durabilities is important especially in the preparation of W.P.C.

by radiation induced polymerizations. It is known that the properties of W.P.C., especially the dimensional stability, hardness, acid-resistancy and alkali-resistancy etc. are improved remarkably comparing those of the

original woods¹⁻⁷⁾. For exact comparison of the degree of improvement in properties the fundamental data concerning the physical properties of the domestic woods are essential. It is also reported that⁸⁻¹⁷⁾ the pure cellulose is degraded to some extents even by 1 Mrad of gamma irradiation giving rise CO₂ and CO from the cellulose but keeps their total crystallinity unchanged. Woods are not pure cellulose, and contain lignin and resin. Since such lignin and resin have aromatic structure, they can absorb and disperse the radiation energy to the whole molecule or they can retard the formation and action of free radicals. Consequently, they make the woods much durable to radiation comparing the pure cellulose^{4, 18, 19)}. The partial degradation of cellulose by radiation will result primarily the decrease of specific gravity and hardness. In this paper the authors describes the changes of specific gravity and hardness of γ -irradiated domestic woods and imported lauans, and discussed their radiation durabilities.

2. Experimental

(1) Materials and Instruments

(a) Wood Samples

Wood stocks of some domestic woods and imported lauans were procured from commercial sources (Table 1). The middle portion of the stocks were cut in tangential direction to the annual ring (Fig. 1), and divided them further. The center of the annual ring is generally not coincides with the geometrical

center of the wood stocks.

The physical properties of woods vary not only with species but also with portions.

Therefore, many samples were used for each wood species so as to minimize the experimental error.

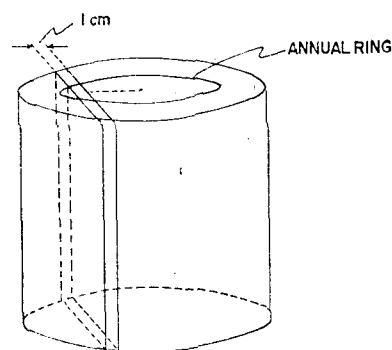


Fig. 1 Sampling from the wood stocks

(b) Radiation source and Tester

Experiments were performed using ⁶⁰Co gamma irradiation facility of K. A. E. R. I. The universal testing machine of Instron Model TML was used in the measurements.

(2) Measurements of Characteristics

(a) Measurements of Water Contents and Water Absorptivity

The wood samples made in the way described in (1), (a) were again cut to 10cm in length, parallel to the stem, and to 1cm×1 cm in section, vertical to the stem. The samples were precisely weighed and dried to constant weight. The weight decreases were expressed as water contents (%). The dried wood samples used in the measurements of water contents were immersed into 30°C

Table 1. Wood samples

Wood species	Origin(home)	Age (approx.)	Wood species	Origin(home)	Age (approx.)
Pinus densiflora	Kyung-Gi Prov., Korea	15	Fraxinus Rhynchophila	Kang-Won Prov., Korea	10
Pinus rigida	"	18	Red lauan	Malaysia	20
Pinus Koraiensis	"	12	White lauan	"	20
Salix Koreansis	"	10	Yellow lauan	"	18
Juniperus Chinensis	"	10	Apitong	"	20
Castanea Crenata	"	14			

water in thermostat to measure the weight increase with time. The weight increases were expressed as the water absorptivity (%) (Table 2).

(b) Measurements of Specific Gravity

The specific gravities were directly calculated from the volume and weight of the wood samples described in (2), (a) (Table 3).

(c) Measurements of Hardness

According to the method described in the ASTM standard²⁰⁾, the hardness was measured for the wood samples described in (2), (a) by using universal testing machine, and expressed in B.H.N. (Brinell hardness number) (Table 4).

The measuring conditions were as following;

Applied Load, $L=40\text{kg}$

Diameter of the ball, $D=7.925\text{mm}$

cross head speed; 0.05 cm/min

(d) Measurements of Bending Strength²⁰⁾

The wood samples made in (1), (a) were cut to 18 cm in length. The bending strength in the normal direction to the annual ring were measured by using testing machine (Table 5). The ball used in the measurements of hardness was detached from the machine and another accessory shown in Fig. 3 was attached to. In the measurement, the constant weight of 40kg was not applied but gradually increased the load until the wood sample is broken. The applied load and the depressed

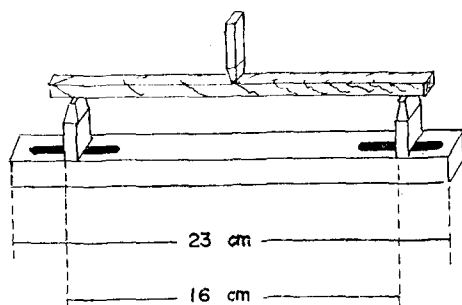


Fig. 3. Sample mounting in the measurements of bending strength

distance at the instant of breakage were read, and they were transformed to the Young's modulus applying the following equation:

$$\text{Young's Modulus } (M) = \frac{Wl^3}{4sa^3b} \text{ (dyne/cm}^2\text{)}$$

(for rectangular bar)

where, W; load

l; distance(cm) between the supporting rods (Fig. 3)

a, b; dimension(cm) in the vertical and horizontal direction

s; flexure(cm)

(e) Measurements of Hardness Changes along the Distance from the Center of the Annual Ring to the Peel

The hardness changes along the distance from the center of the annual ring to the peel were measured in both normal and tangential direction to the annual ring. The measuring conditions were the same as that of the measurements of hardness in normal direction to the annual ring, described in (2), (c) (Fig. 4-1, 4-2),

(f) Measurements of Resin Contents

The resin in the wood samples was extracted by n-hexane in Soxhlet extractor. The weight of the resin was precisely weighed after evaporation of the solvent, and the resin contents were expressed in weight % (Table 6).

(3) The Changes of Physical Properties of Woods by gamma Irradiation

(a) Measurements of Dose Rates

According to the method reported by H. Gladys Swope²¹⁾ the chemical dosimetry was performed utilizing the oxidation of ferrous ion in ferrous sulfate solution.

(b) Changes of Specific Gravity

The changes of specific gravity were measured for the wood samples described in (2), (a). The dose rate was fixed to 2.54×10^5 rad/hr. (Fig. 5).

(c) Changes of Hardness

Table 2. Water contents and water absorptivity of various wood in weight percent

Woods	Water Contents	Duration of Soaking in 30°C Water(hrs.)									
		0.7	1.5	17	24	48	72	140	188	262	432
populus deltoides	10.9	26.3	32.5	75.5	86.7	112.8	131.2	172.7	183.9	197.5	218.9
pinus rigida	12.3	20.1	23.3	40.2	45.7	56.7	64.9	91.0	97.9	105.6	121.9
pinus densiflora	12.3	23.3	26.2	41.1	45.8	54.4	66.7	87.3	92.0	98.3	109.1
red lauan	10.7	9.1	11.2	31.0	36.9	48.0	55.9	77.5	84.2	94.0	109.5
white lauan	11.2	11.7	13.3	32.5	38.0	48.4	56.2	76.8	82.5	91.7	105.4
yellow lauan	11.4	8.1	10.2	29.8	34.4	43.4	51.4	69.9	76.7	84.8	100.4
apitong	11.2	5.1	6.8	22.1	25.8	32.0	37.0	47.9	50.7	55.2	61.3

Table 3. Specific gravity of woods

Woods	pinus densiflora	pinus rigida	populus deltoides	yellow lauan	white lauan	red lauan	apitong
sp. gr.	0.55	0.42	0.39	0.54	0.43	0.65	0.74

Table 4. The hardness of woods

Woods	pinus densiflora	pinus rigida	populus deltoides	yellow lauan	white lauan	red lauan	apitong
Hardness* (B. H. N.)	2.56	1.20	1.10	1.80	1.70	2.50	2.60

* measured in normal direction to the annual ring.

Table 5. The Bending strength of woods*

Wood	pinus densiflora	pinus rigida	populus deltoides	yellow lauan	white lauan	red lauan	apitong
Weight of load(kg)	30.0	17.0	18.7	34.0	25.0	44.0	45.0
Young's modulus (dyne/cm ²)	3.1×10 ⁷	2.2×10 ⁷	2.5×10 ⁷	5.6×10 ⁷	3.6×10 ⁷	8×10 ⁷	1×10 ⁸

* measured in normal direction to the annual rings

Table 6. Resin contents in woods

Woods	pinus densiflora	populus deltoides	lauans	Salix Koreansis	Juniperus Chinensis	Castanea Crenata	pinus koraiensis
Resin Contents in Weight %	0.95	0.35	0.76	2.10	2.10	0.55	1.51

The changes of hardness in normal direction to the annual ring were measured. The irradiation conditions were the same as that described in (2), (b). The changes were expressed in % based upon the B.H.N. of non irradiated samples. (Fig. 6).

3. Results and Discussion

(1) Characteristics of Various Woods

As Table 2 shows, the water contents of woods dried for long time at room temperature is 10-12% regardless the species. However,

the rate and amounts of water absorption is quite different from wood to wood. *Populus deltoides* absorbs water remarkably showing 2 times of its weight. Other woods such as *pinus densiflora*, *pinus rigida* and *lauans* absorb moderate amounts of water showing 100% of its weight. *Apitong* absorbs less amounts of water showing only 50% of its weight. It was confirmed that the rate of water absorptivity is proportional to the maximum absorption amount of water.

As Table 3 shows, the specific gravity is related not only to the density of wood cellulose but also to the components of resin in wood.

The values are larger for *apitong*, red *lauan* and *pinus densiflora* than those for *pinus rigida* and *populus deltoides*.

As Table 4 shows, the hardness of woods in normal direction is generally high for the woods of high specific gravity such as *apitong* and red *lauan*. The order of hardness is *apitong* > red *lauan* > yellow *lauan* > white *lauan* > *pinus densiflora* > *pinus rigida* > *populus deltoides*. *Populus deltoides* has the lowest values of both specific gravity and hardness. In the measurements of hardness in normal direction, as we see in Fig. 1 and in the description of (1), (a), only the central part of the wood sample can precisely be measured in normal direction to the annual ring. If the hardness is measured in this way for the outer part (peel part) of the wood sample it becomes no more the hardness in normal direction but that of tangential direction. It may be said that the hardness is proportional to the specific gravity even though there are some exceptions (Fig. 2). Therefore, sp. gr. may be a criterion in the selection of soft or hard wood.

Apitong has high value of bending strength, 45kg. The order of bending strength is *apitong* > red *lauan* > yellow *lauan* > *pinus den-*

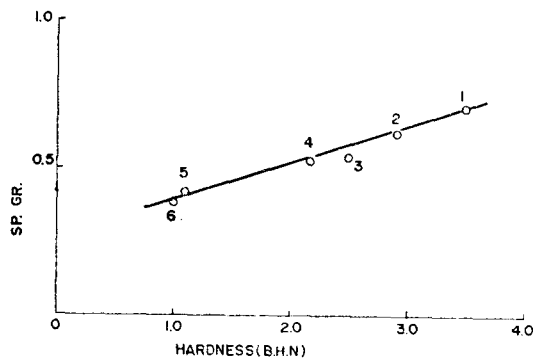


Fig. 2. Hardness v. s. specific gravity of woods

1. *Fraxinus rhynchophylla*,
2. *Cornus controversa*
3. *Pinus densiflora*, 4. *Juniperus Chinensis*,
5. *Pinus rigida*, 6. *Populus deltoides*

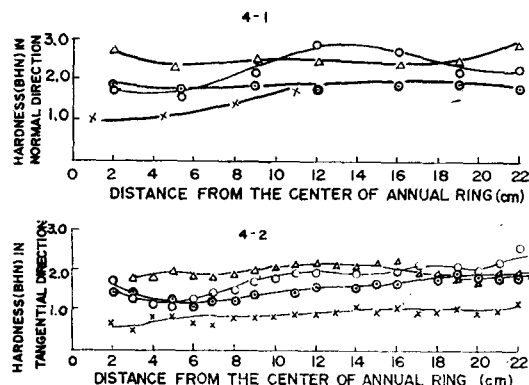


Fig. 4. Variation of Hardness from the center of annual rings

4-1; hardness in normal direction to the annual rings

4-2; hardness in tangential direction to the annual rings

△; *apitong*, ○; red *lauan*, ⊙; yellow *lauan*,
×; white *lauan*

siflora > white *lauan* > *populus deltoides*. This order is roughly consistent with that of hardness, considering that the inaccuracy in the measurements of hardness in normal direction. Fig. 4-1 and 4-2 show the changes of hardness along with the distance from the center of the annual ring to the peel. As the Figures show, it is general trend that the changes of hardness are extreme in the central

and outer parts of the wood stocks, but not much in the middle part. The hardness of the middle part is getting larger with going out to the peel part. It is certain that the heart wood is generally not hard but there are some hard points. The lower hardness in outer part may be attributable mainly to the partial deterioration of wood cellulose during storage. The lower resin contents and softness of the wood cellulose in the outer part of the wood may also be other causes.

It is noteworthy that the hardness is changed significantly with the direction of measurement even the wood species is the same. Generally, the hardness values measured in the normal direction to the annual ring are significantly larger than those measured in tangential direction. However, the reproducibilities of the data are better in tangential direction. Thus, to get experimental data, the measurement in tangential direction is recommendable.

As Table 6 shows, *Juniperus Chinensis* contains a lot of resin. Plenty of resin is also contained in *Salix Koreansis* and *Pinus Koraiensis* etc. *Populus deltoides*, *Castanea Crenata*, *lauans*, and *pinus densiflora* etc.

contain less resin.

The *Salix Koreansis* contains much resin but the hardness is low. Such a phenomenon may due to the softness of the contained resin. Therefore, the woods of high resin contents are not always the woods of high specific gravity, or hard woods.

(2) Changes of Physical Properties of Woods by gamma Irradiation(Radiation Durabilities)

The studies on the changes of physical or mechanical properties of woods were carried out in some foreign countries using Beech, pine⁶⁾, and oak⁷⁾ etc., all woods produced in their own countries. Such a study performed for the usually available woods in this country is hardly found. It was reported that the cellulose is readily degraded its main chain by radiation⁸⁻¹⁷⁾, and further degraded to evolve hydrogen, carbon monoxide and carbon dioxide etc^{11, 13, 14, 17)}. Thus it is expected that the specific gravity may be varied by radiation. However, as Fig. 5 shows, the specific gravity is not decreased. There is rather a tendency of slight increase under the irradiation of 108 Mrad with the dose rate of 2.54×10^5 rad/hr. We consider the reasons as following; 1) wood is not pure cellulose and contains lignin and resin of aromatic struc-

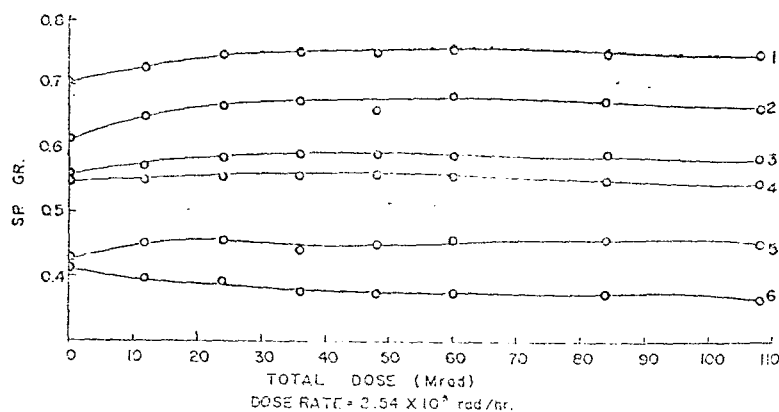


Fig. 5. The effect of gamma irradiation on specific gravity of woods
1. *Fraxius rhynchophilla*, 2. *Cornus Controversa*, 3. *Pinus densiflora*,
4. *Juniperus Chinensis*, 5. *Pinus rigida*, 6. *Populus deltoides*

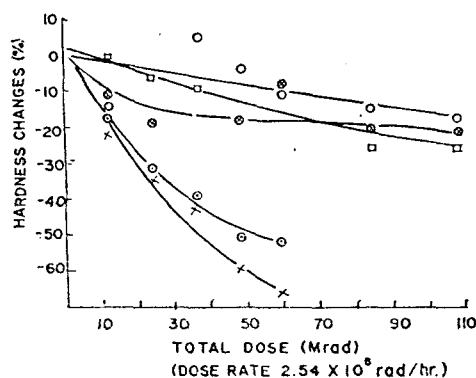


Fig. 6. The effect of gamma irradiation on the hardness of woods

○; pinus densiflora, ×; pinus rigida,
 ⊙; populus deltoides, ⊗; Juniperus Chinensis,
 □; Fraxinus rhynchophylla

tures. The lignin and resin in wood can certainly protect the radiation degradation of homocellulose to some degree, and consequently no degradation occurred under such a dose¹⁸⁾. 2) the dried wood samples absorbed some moisture of the air during irradiation, 3) the free radical formed by gamma irradiation, 3) the free radical formed by gamma irradiation reacted with oxygen in the air forming peroxide etc.

Considering the fact that the corresponding degradation was occurred to result hardness decrease, the possible explanation by 1) is not reasonable and should be excluded. Siau *et al*²²⁾ reported that the irradiated woods have hygroscopic character. On the other hand, Arthur *et al*²³⁾ reported that when the cotton cellulose is irradiated the change of hygroscopic character is not much, and the gas evolution rate is smaller than the oxygen decreasing rate due to oxidation reaction. Therefore, the explanation by 2) and 3) may be reasonable, but which one of the two is more significant is still under study.

The data obtained in the measurements of hardness change by gamma irradiation (Fig.

6) show that the hardness is generally decreased with irradiation, which is consistent with the fact previously reported. Among them, the hardness decrease in soft woods such as pinus rigida and populus deltoides is especially extreme. Pinus rigida was so easily ruptured in 84 Mrad irradiation that the hardness can hardly be measured. It was reported that woods are ruptured by 200 Mrad of gamma irradiation²⁴⁾. This is much higher value than our data. Generally, the degree of radiation rupture was severe for soft woods or woods of lower resin contents. In this point of view, it may be said that pinus rigida and populus deltoides are not suitable as raw materials for the preparation of W.P.C. But, practically, in the preparation of W.P.C. the dose required is not so much, and usually 10 Mrad is sufficient. Further, the wood is irradiated with the impregnated monomer which can play a role of absorbent of radiation energy. Therefore, it is certain that even soft woods are sufficiently useable under such circumstances for preparation of W.P.C.

4. Conclusions

(1) The hardness of wood is generally proportional to the specific gravity and to the bending strength.

(2) The hardness of wood measured in tangential direction to the annual ring increases along with the distance from the center of the annual ring to the peel.

(3) The changes of specific gravity under 100Mrad of gamma irradiation are not remarkable.

(4) The hardnesses of soft woods are decreased more rapidly than those of the hard woods, but the decreases of hardnesses are not extreme upto 10Mrad suggesting that the woods are sufficiently durable to gamma irradiation of below 10Mrad in the preparation of W.P.C.

References

- 1) Kent, J.A. *et al*; Preparation of Wood-Plastic Combinations using gamma Irradiation to induce Polymeirzation Proc. Int. Symp. Rad. Ind. Polymerization and Graft Polymerization, 29-30 November, 1962. Battele Memorial Inst. U.S. A.E.C. TID 7643, 335-344 (1962)
- 2) Siau, J.F. *et al*; Wood-Plastic Combniations Using Radiation Technique. Forest Prod. J. 15, 260 (1965) through T. Czvikovzky; Wood-Plastic Combination by Monomer Impregnation and Radiation Polymerization, Atomic Energy Rev. 5(3), 3 (1968)
- 3) Kenaga, D.L. *et. al*; Radiation Graftings of Vinyl Monomers to wood, *ibid.* 12, 161 (1962) through T. Czvikovzky; Wood-Plastic Combination by Monomer Impregnation and Radiation, polymerization Atomic Energy Rev. 6(3), 3 (1968)
- 4) Ramalingam, K.V. *et al*; Radiation Induced Graft Polymerization of Styrene in Wood. Proc. 4th Cellulose Conf. 81-19 October, 1962, Interscience Publishers, New York, J. Polym. Sci. C2, 153 (1963)
- 5) Kent, J.A. *et al*; Manufacture of Wood-Plastic Combinations by Use of gamma Radiation, Industrial Use of Large Radiation Sources(Proc. Conf. Salzburg, 1963) 1, 377-404, I.A.E.A., Vienna (1963)
- 6) Karpov, V.L. *et al*; Radiation makes Better Woods and Copolymers, Nucleonics, 18(3), 88 (1960)
- 7) Czvikovszky, T.; Wood-Plastic Combinntions by Monomer Impregnation and Radiation Polymerization. Rep. No. 8 of the Plastic Research Inst. (1967) through Cziakovovzky; Wood-Plastic Combination by Monomer Impregnation and Radiation Polymerization, Atomic Enery Rev. 6(3), 3 (1968)
- 8) Shoenfle, C.S. and Connel, L.A.; Effect of Cathod Rays on Hydrocarbzn Oils and Paper. Ind. Eng. Chem. 21(6) 529-537 (1929) through Isotopes and Rad. Tech. Sec. 5, 3(3), 326 (1966)
- 9) Seaman, J.F. *et al*; Effect of High Energy Cathod Rays on Cellulose. *ibid.* 44(12), 2848-2852 (1952) through Isotopes and Rad. Tech. Sec. 5, 3(3), 326 (1966)
- 10) Charlesby, A.; Degradation of Cellulose by Ionizing Radiation. J. Polym. Sci. 263-270 (1955)
- 11) Glegg, R.E. and Kertesz, Z.I.; Effect of Gamma Irradiation on Cellulose. J. Polym. gci. 26, 289-297 (1957)
- 12) Glegg, R.E.; The Influences of Oxygen and Water on the After-Effect in Cellulose Degradation by gamma Rays. Ran. Res. 6, 469-473 (1957)
- 13) Blouin, F.A. and Arther, J.C.; The Effect of Gamma Radiation. (I). Some of the Properties of Purified Cotton Irradiated in Oxygen and Nitrogen Atmosphere. Textile Res. J. 28, 198-204 (1958) through Isotopes and Rad. Tech. Sec. 5, 3(3), 326 (1966)
- 14) Arther, J.C.; Proposed Mechanism of the Effect of High Energy Gamma Radiation on Some of the Molecular Properties of Purified Cotton. Textile Res. J. 28, 204-217 (1968)
- 15) Blouin, F.A. and Arther, C.J.; Degradation of Cotton in Oxygen Atmosphere by Gamma Radiation. J. Chem. Eng. Data 5, 470-475 (1960)
- 16) Arther, C.J. *et al*; The Effect of Gamma Radiation on Cotton Cellulose. Amer. Dyestuff Reprtr. 49, 383-388 (1960)
- 17) Huang, R.Y.M.; Radiation Induced Graft Copolymers of Cellulose. Ph D. thesis, Univ. of Toronto (1963) through Isotopes and Radiation Tech. Sec. 5, 3(3) (1963)
- 18) Smith, D.M. and Mixer, R.Y.; The Effect of Lignin on the Degradation of Wood by Gamma Irradiation. Radiation Res. 11, 776-780 (1959)
- 19) Fuccillo, D.A. *et al*; Recent Development in the Production of Wood-Plastic Combinations. Technical and Economic Considerations for an Irradiated Wood Plastic Materials. Isotope and Rad. Tech. 3(2). 115-146 (1965)
- 20) ASTM Standard (1958) part 6, D 143-52, American Society for Testing Materials, Philadelphia. Pa. U.S.A.
- 21) Gladys Swope, H.; ANL Report, USAEC Contract No. W-31-109eng-39 (1958)

- 22) Siau, F. J. *et al*; A Review of Developments in Dimensional Stabilization of Wood using Radiation Technique. *Forest Prod. J.* 15(4), 162-166 (1965) through *Isotopes and Rad. Tech.* 3(2), 120, Winter (1965-1966)
- 23) Arther, C. J. and Blouin, F. A.; Radiation Induced Graft Polymers of Cellulose. *Proc. Int. Symp. Rad. Polymerization and Graft Polymerization*, 29-30, November, 1962. Battle Memorial Inst., USAEC TID 7643, 319-334 (1962)
- 24) Burmester, A.; Zur Vergütung von Holz durch Strahlenpolymerisierte Kunststoff-Monomere, *Holzroh-U, Werkstoff* 25, 11 (1967) through *Atomic Energy Rev.* 6(3), 8 (1968)