

## CHAPTER III

### LITERATURE REVIEW

In recent year, natural fiber reinforced in thermoplastics has been increasingly well accepted because it can be more easily processed than other rigid fibers such as fiberglass, mica, etc. Therefore, natural fiber has an increased share on the market of reinforced plastics and filled polymers. The popular natural fiber used as reinforcement in thermoplastic composite was the wood sawdust because it can be obtained in large quantity as waste material from the sawmill factory. This will clearly result in a value-added product by utilizing waste wood.

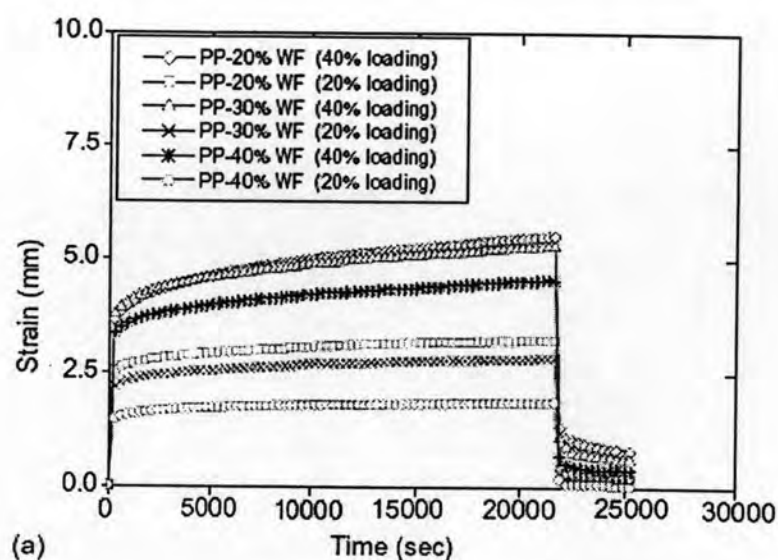
Stark and Berger (1997) studied the influence of type and concentration of woodflour on the mechanical properties of wood filled polypropylene (PP) composites. They used five species of woodflour, namely; ponderosa pine, loblolly pine, maple, and oak as fillers for polypropylene matrix at various compositions of 20, 30, 40, 50, and 60% by weight. The results of this experiment indicated the mechanical properties that were shown in Table 3.1. From this table, it is important for the end user to be aware of the differences that species variations may have on their product, because species can be regionally influenced. In general, the hardwoods exhibited slightly better tensile and flexural properties than did the softwoods. In addition, their results showed that with increasing woodflour content, flexural and tensile modulus, and notched impact energy increased, while flexural and tensile strength, tensile elongation, and unnotched impact energy decreased.

The other property that is important to wood composites is creep behavior. Because the wood composite was deformed when was used as a long time or high temperature. Creep behavior of wood composites was studied by Lee et al. (2004) in the system of polypropylene wood composite. The specimens used for creep test were dog bone shape. The value of applied stress for creep test was chosen at 40% of

tensile strength of polypropylene. The time period used for the creep test was kept constant at 6 hr. The authors reported that the creep strain decreased as the woodflour level increase as shown in Figure 3.1.

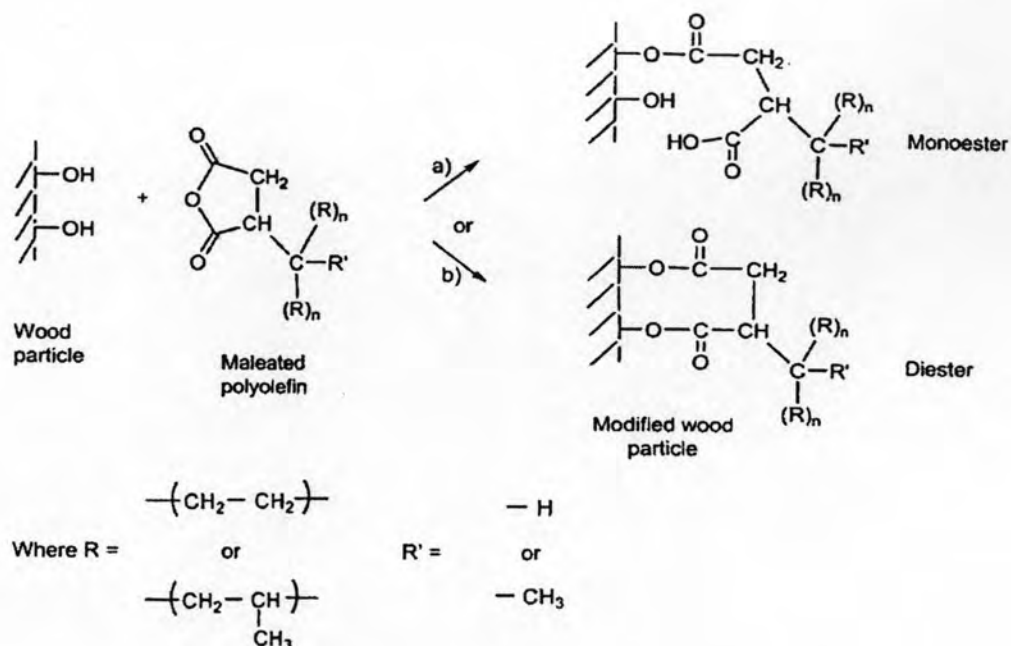
**Table 3.1:** Summary of mechanical properties of several species of woodflour filled polypropylene (Stark and Berger, 1997).

Material	Izod impact		Tensile properties			Flexural properties	
	Notched	Unnotched	Strength	Elongation	Modulus	Strength	Modulus
	(J/m)	(J/m)	(MPa)	(%)	(GPa)	(MPa)	(GPa)
PND-20	15.4	128	26.5	5.65	1.99	41.6	1.89
PND-30	19.0	95	24.6	3.12	3.24	43.1	2.58
PND-40	20.8	76	25.5	2.29	3.71	44.2	3.22
PND-50	20.5	58	23.0	1.67	4.25	41.8	3.66
PND-60	21.1	41	20.1	1.44	4.56	38.8	4.04
LBL-20	12.4	120	24.9	4.83	2.14	40.6	1.71
LBL-30	12.7	75	23.7	3.45	2.53	41.2	2.28
LBL-40	13.7	49	21.4	1.76	3.28	39.3	2.84
LBL-50	13.8	42	19.7	1.32	3.97	37.1	3.40
LBL-60	10.4	31	17.8	0.92	4.32	34.1	3.81
MPL-20	12.8	113	27.9	4.11	2.87	46.2	2.00
MPL-30	15.0	87	27.1	3.26	3.33	46.5	2.47
MPL-40	16.5	63	25.6	2.03	4.72	45.4	3.23
MPL-50	17.7	49	24.0	1.44	5.20	42.1	4.16
MPL-60	17.9	44	19.9	1.13	4.77	38.0	4.35
OAK-20	14.6	87	27.2	4.66	2.48	44.1	1.83
OAK-30	17.5	63	25.7	2.43	3.81	45.9	2.87
OAK-40	18.6	68	25.2	2.09	4.19	44.8	3.39
OAK-50	20.9	46	23.4	1.51	4.79	42.8	3.99
OAK-60	18.8	33	19.8	1.20	5.05	38.1	4.60
PP	15	600	28.5	10.44	1.31	34.7	1.03



**Figure 3.1:** The effects of stress level on creep behavior of WPC (Lee et al., 2004).

The use of woodflour as a reinforcement in many thermoplastics creates some drawbacks including a lack of a good interfacial adhesion and poor resistance to humidity absorption since natural woodflour is hydrophilic in nature while thermoplastic matrix is rather hydrophobic (Albano et al., 2001; Albano et al., 2002). In 1968, Meyer was possibly the first person who suggested the use of a coupling agent which called a crosslinking agent to improve the interfacial adhesion between the woodflour fillers and the thermoplastic matrix. Later on, many researchers were trying to further improve the properties of wood composites. The improvement of wood composites properties includes the methods such as treatment with silane or adding compatibilizer such as acrylic acid, acrylic esters, maleic anhydride, etc. However, the major measure to enhance an interfacial adhesion between woodflour filler and polypropylene matrix was by adding the compatibilizer e.g. maleic anhydride graft polypropylene (MA-g-PP) because technique is easier and faster than silane treatment. The effect of a compatibilizer on interfacial property enhancement in wood composite was from an esterification reaction between hydroxyl groups of wood particles and anhydride group of MA-g-PP as illustrated in Figure 3.2.



**Figure 3.2:** Modification scheme for esterification reaction between wood particles and maleated polyolefins: monoester (a) and diester (b) formation (Carlborn et al., 2005).

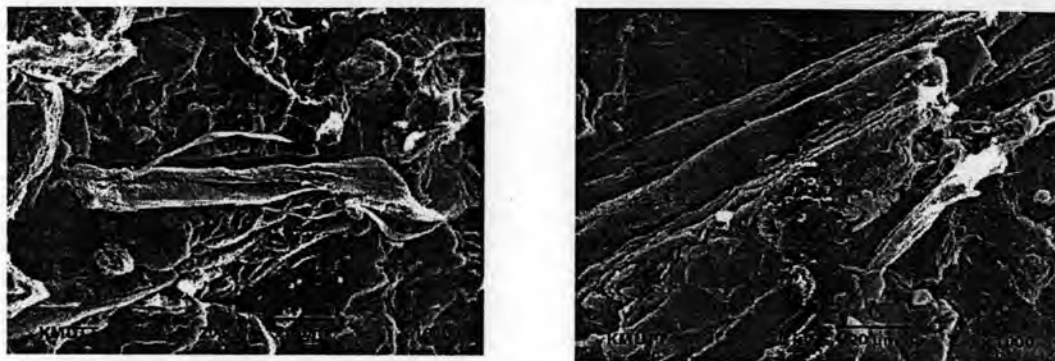
Based on the work of Qiu et al. (2005), the authors used maleated anhydride graft polypropylene (PP-g-MA) as a compatibilizer in their wood composites that consisted of isotactic polypropylene and fibrous cellulose. In this system, the isotactic polypropylene was mixed with 30% by weight of fibrous cellulose (50-350 microns in length, ~20 micron in diameter). From the results, they found that the compatibilizer of about 10% by weight of the fibrous cellulose could improve the interfacial adhesion of the wood composites resulting in an enhancement of the composite mechanical properties. As aforementioned, an addition of PP-g-MA into wood composite system can generate esterification reaction between PP-g-MA and hydroxyl groups of cellulose. The mechanical property enhancement in this fibrous cellulose composite was evidently attributed to the strong interfacial adhesion between filler and polymer matrix. The wood composite treated with the optimal concentration of PP-g-MA of about 10% by weight of fibrous cellulose was found to

increase tensile strength and Young's modulus of the composite from 31.7 MPa to 33.7 MPa and 0.51 GPa to 0.95 GPa, respectively.

The effect of the contents and types of PP-g-MA on mechanical properties of wood composites was also studied by Sombatsompop et al. (2004-2005). The particles sizes of wood sawdust used were in the ranges of 100-300 microns. The contents of wood sawdust particles added into the PP matrix were ranged from 0 to 30% by weight. The results demonstrated that addition of 30% by weight of woodflour into polypropylene matrix was found to enhance the tensile modulus from 830 MPa to 1200 MPa and was found to decrease the elongation at break from 21% to 5.5% compared with those of polypropylene ( $E = 830$  MPa,  $\epsilon = 21\%$ ). At the same composition of woodflour, the tensile strength decreased from 35 MPa of the polypropylene matrix to 27 MPa. The decrease of tensile strength was probably caused by poor dispersion of the fibers in the matrix, moisture pick-up, and an increase of interfacial defects between the polypropylene matrix and the untreated fibers. In addition, the interfacial adhesion improvement of the wood composite was studied using three types of PP-g-MA compatibilizers, namely; MZ203D, MD353D, and MD411D. They found that the excess concentrations of PP-g-MA of approximately 4.2% to 11.1% by weight of woodflour showed marginal improvement of the mechanical properties. Furthermore, the optimal concentration of PP-g-MA was found to be 2% by weight of woodflour resulted in the maximum tensile modulus and tensile strength of the composites. The interfacial interaction improvement of this composite was confirmed by scanning electron microscope (SEM) as shown in Figure 3.3.

However, the obvious disadvantages of using a compatibilizer to improve interfacial adhesion for thermoplastic wood composites are cost of the chemicals and the need to determine optimal concentration of the compatibilizer in each woodflour-filled composite. Therefore, the technique based on an irradiation of composites appears as an alternative to compatibilizing of wood composites. Using irradiation to improve the filler/matrix interfacial adhesion is cheaper and easier to process than using a compatibilizer or treating with silane. This is the only technique that

introduces energy into a material to generate favorable changes in thermoplastic wood composite structure, provided it was used in a proper dose and under an appropriate condition.

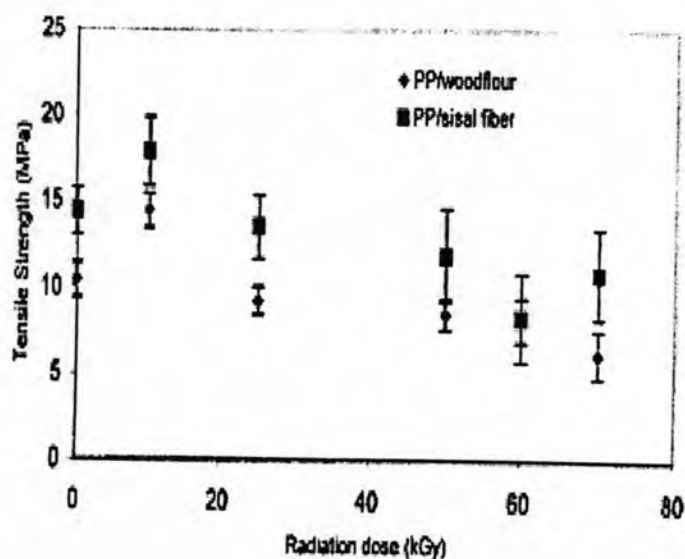


**Figure 3.3:** SEM micrographs of PP/wood composites: (a) untreated, (b) treated with 2% by weight PP-g-MA (Sombatsompop et al., 2004).

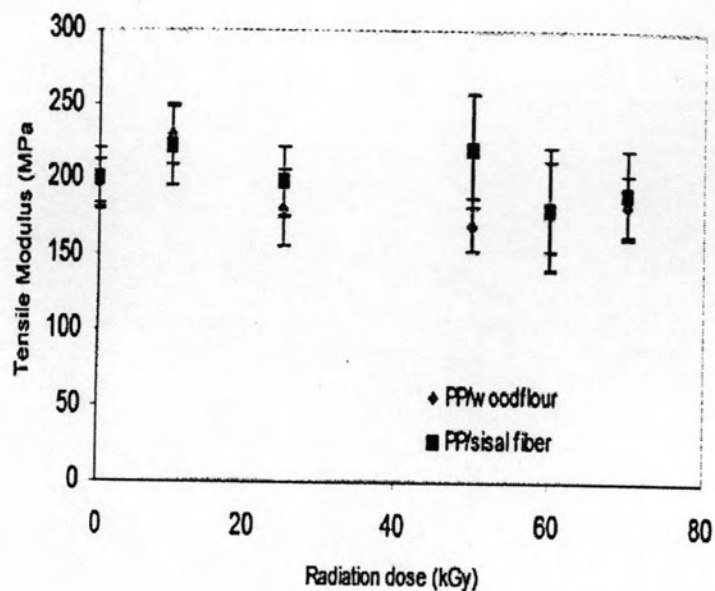
Recently, the effect of gamma ray on mechanical properties of polypropylene was studied by Reyes et al. (2001). They used various doses of gamma irradiation ranging from 1 to 7 MRads at a constant dose rate of 0.48 MRads/hr, in oxygen atmosphere. The results obtained revealed that low irradiation in a range of 1 to 5 MRads can improve the mechanical properties of polypropylene substantially. Compared with unirradiated polypropylene, young's modulus and tensile strength of the irradiated polypropylene was found to significantly increase from 295 MPa to 605 MPa and from 33 MPa to 34.5 MPa, respectively. These properties decreased with greater than 5 MRads radiation dose as tended to polypropylene degrade in chain scission manner at high radiation level.

Albano et.al. (2001-2001) studied influence of gamma irradiation on the properties of polypropylene wood composites filled with 40% by weight of woodflour. The composites were irradiated with gamma rays at radiation doses of 10, 25, 50, 60 and 70 kGy using a fixed dose rate of 0.48 kGy/hr in air atmosphere. Figures 3.4-3.6 show the values of tensile strength, tensile modulus, and elongation at

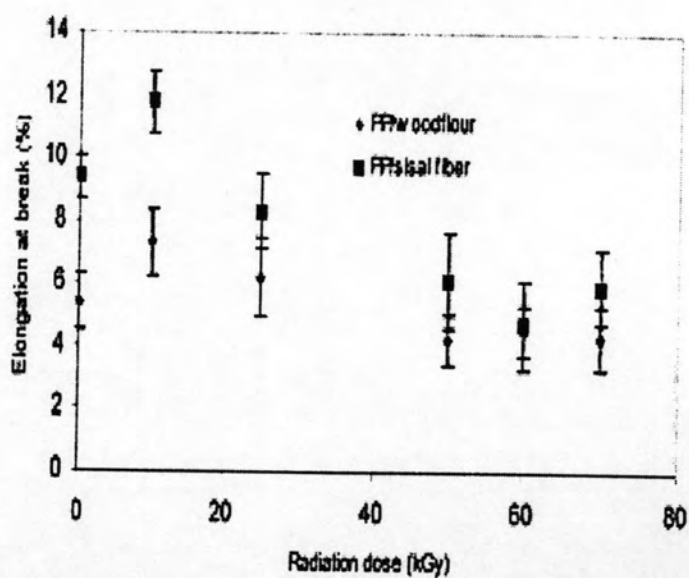
break of the polypropylene wood composites. The mechanical property enhancement in the polypropylene composite irradiated by 10 kGy of gamma radiation was attributed to several reasons including; a slight cross-linking which may be experienced by polypropylene, the higher polymer-filler interaction due to increased hydrophilicity of polypropylene when it is irradiated, which is due to the functional OH groups. In addition, the filler has high concentration of OH groups which may signify interactions of the hydrogen bridge type between the filler and polypropylene. However, the effect of gamma irradiation on the DSC thermogram shown in Figure 3.7 suggested constant melting temperatures at low radiation dose and tended decrease at higher dose. This effect was possibly caused by additional oxidative degradation of polypropylene when the polymer received too high radiation dose.



**Figure 3.4:** Tensile strength of PP/wood and sisal fiber composite vs. irradiation dose (Albano et al., 2002).

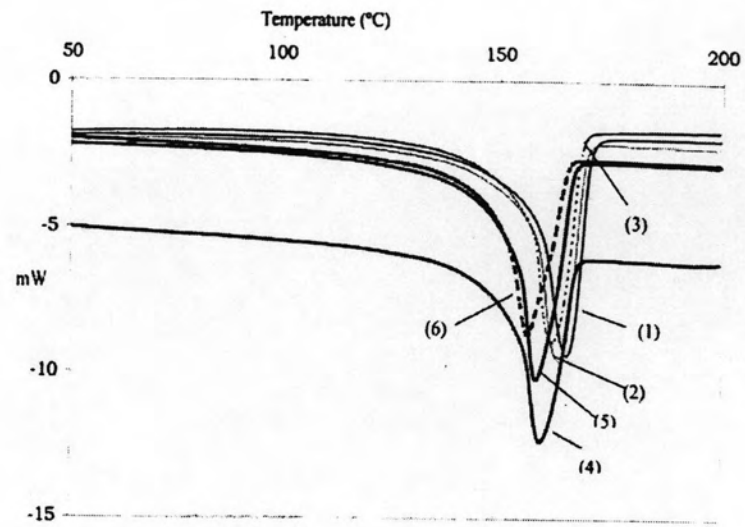


**Figure 3.5:** Tensile modulus of PP/wood flour and PP/sisal fibre composites vs. irradiation dose (Albano et al., 2002).



**Figure 3.6:** Elongation at break of PP/wood and sisal fiber composite vs. irradiation dose (Albano et al., 2002).





**Figure 3.7:** The effect of  $\gamma$ -irradiation on the DSC thermograms (second heating) of PP/wood flour: (1) unirradiated, and irradiated: (2) 10 kGy, (3) 25 kGy, (4) 50 kGy, (5) 60 kGy, (6) 70 kGy (Albano et al., 2002).