PROPERTIES OF TRANSFER-MOLDED WOOD-FIBER/POLYSTYRENE COMPOSITES

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ABSTRACT

Transfer-molded composites combining polystyrene, wood particles, and three bi-functional coupling agents were prepared and evaluated for physical and mechanical properties. Pure 685D polystyrene (PS) (75–100% by weight) was combined with 100-mesh (0.15-mm sieve opening) particles prepared from thermomechanically pulped quaking aspen (*Populus tremuloides*) (0–25% by weight). Three coupling agents, polystyrene/poly(methacrylic) (both low and high molecular weight) and polystyrene/poly(vinyl acetate) developed at Michigan Technological University, were added in an effort to promote compatibility between the hygroscopic wood fiber and the non-polar hydrophobic polystyrene. Mechanical tensile testing was used to assess the respective composite's tensile elastic modulus and tensile strength. A polystyrene/poly(methacrylic) acid (PS-PMAA) coupling agent was found to be the most effective with regard to enhanced tensile elastic modulus at higher fiber-loading levels (enhancement levels of 11.3–23.8% over pure PS). A fiber/PS composite using low molecular weight PMAA (PS-PMAAL) as a coupling agent demonstrated the best tensile strength retention characteristics at higher fiber-loading levels. Initial results show high variability in material properties over the range of fiber-loading levels, and between coupling agent type. It is clear, however, that certain coupling agents do have a positive effect on composite properties.

Keywords: Composite, coupling agent, mechanical properties, polystyrene, tensile strength, transfer molding, wood fiber.

INTRODUCTION

The recycling of municipal wastes is becoming an increasingly important issue as new landfill sites become scarce, and the threat to the environment, caused by overflowing debris, grows. The single largest stream of municipal wastes is that of wood-based materials; it is in excess of 40% of all wastes. The second major segment is plastics, which make up 10% by weight, or 30% by volume, of all landfill

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TABLE 1. *Physical properties of block components.*

Property/condition	PS	P (vinyl acetate)	P (methacrylic acid)
η_0@180 C, N-s/m[2]	14,800[1]		
η_0@200 C, N-s/m[2]		200[2]	24,950[3]
η_0@220 C, N-s/m[2]	5,000[2]		
γ@200 C, 10^{-3} N/mm		2.59[4]	2.94[3]
γ@200 C, 10^{-3} N/mm	2.78[4]		

Source: [1] Bird et al. 1987; [2] Van Krevelen and Hoftyzer 1976; [3] Van Krevelen and Hoftyzer 1976 (the viscosity and surface tension of poly(methyacrylic acid) at 200 C was obtained from group contribution methods); [4] Brandup and Immergut 1989.

materials (U.S. Congress, Office of Technology Assessment 1989). Among major plastics, polystyrene (PS) has been extensively used for packaging, containers, and appliances, and in many other disposable products. These products often end up in landfills. One potential solution for utilizing these recycled materials is to convert them into recyclable and reusable composites, which would be composed primarily of wood fiber and thermoplastic polymers.

Wood fiber possesses a number of potential advantages as a suitable candidate in fiber-reinforced polymer composites. Among these advantages, those of major importance include low price, low density, low abrasiveness, and the absence of a potential health hazard during processing (Sean et al. 1990).

Unlike PS, which is a non-polar hydrophobic material, wood fiber is a polar substance primarily due to the presence of the hydroxyl groups in its constituent polymers (i.e., cellulose, hemicelluloses, and lignin). This leads to poor compatibility between the two types of material and is disadvantageous with regard to mechanical properties and dimensional stability. This effect has been reported by Hon et al. (1992). It was found that newspaper fiber, coated with plastic without the use of a coupling agent, protruded from the fracture surface of the polymer matrix.

In order to promote the chemical affinity between wood fiber and PS, the use of a coupling agent is beneficial. Maldas and Kokta (1989, 1990a, b, c) researched several types of coupling agents or chemical treatment, e.g., phthalic anhydride, isocyanate, and inorganic salt/polyvinyl chloride.

Bonding mechanisms between wood, polystyrene, and coupling agents

Interfacial bonding in wood-fiber/polystyrene composite materials offers challenges that may be partially overcome with the use of block copolymer coupling agents. This class of coupling agent covalently bonds or interacts with both the wood fibers and the polystyrene matrix. It was expected that vinyl acetate (VAc) or methacrylic acid (MAA) blocks of the copolymer coupling agents used in this study would bond with the wood fibers, while the polystyrene blocks will become an intimate part of the base polystyrene resin.

A number of factors affect coupling agent effectiveness. To promote good surface coverage, a coupling agent should have a combination of relatively low viscosity and interfacial tension. Table 1 gives an indication of the zero-shear viscosities (η_0) and surface tensions (γ) of the block components that make up the coupling agents used. Coupling agent interfacial density is dependent on the molecular weights and molecular weight distributions of the blocks. If the molecular weight of the wood-interacting block is relatively low, more coupling-agent molecules will be found in the interface. In this case the molecules are not considered to be as well-anchored as those with higher molecular weights. The molecular weight of the resin-interacting block also has an effect on the molecular density of the coupling agent at the interface. High molecular weights provide the necessary entanglement sites within the base resin. At very high molecular weights, however, a lower molecular density of interfacial active material (coupling

TABLE 2. *Polystyrene (685D Dow Chemical) specifications.*

Melt flow rate	$2.7 \cdot 10^{-3}$ g/sec
Vicat softening point	107.7 C
Specific gravity	1.04
Tensile strength	56.54 MPa
Tensile elastic modulus	3.34 GPa

agent) is thermodynamically favored. Concave surfaces require a higher PS-block molecular weight to achieve high coupling agent interfacial concentration (Meier 1987). Conversely, convex wood surfaces will require block components of lower molecular weight.

Block copolymer coupling agents were added to the wood and polystyrene system in an effort to improve the strength and stiffness. Other factors that could influence the properties may be concentration and chemical structure of the coupling agent, fiber loading (Maldas and Kokta 1991), and various processing variables (Sean et al. 1991). By considering the results of previous studies, the following objectives were formulated:

1) Produce a wood-fiber/polystyrene composite using three types of coupling agent and compare the bonding effectiveness among the respective coupling agent types.

2) Determine the effect of coupling agents and wood-fiber content on the tensile properties of wood-fiber/polystyrene composites.

MATERIALS AND METHODS

Preparation of wood-fiber PS mixture

Raw materials used in this experiment were quaking aspen fiber (*Populus tremuloides*), prepared by thermomechanical pulping (TMP), and a thermoplastic polystyrene (685D Dow Chemical). The commercially produced aspen fiber was screened to 100-mesh size (0.15-mm sieve opening) for mixing with the PS. The physical and mechanical properties of pure 685D PS are reported in Table 2.

Prior to mixing the fiber and polystyrene, the fiber was first dried to a moisture content of 3% (oven-dry weight). The dry, finely ground coupling agent powders were mixed with wood fiber to achieve an even consistency. The PS pellets were then added to the wood-fiber/coupling agent mix.

The blending of the wood-fiber PS mixture was accomplished by a Brabender "Twin Screw Mixer" (TSM). The TSM was used in conjunction with a "Prep Center," which is a completely enclosed small batch preparation unit, operating at a constant torque and at variable or fixed speeds. The two counter-rotating intermeshing screws in the TSM provide uniform blending for the raw material through a high shearing action, thus producing a consistent distribution of wood fiber throughout the PS matrix. The temperature was maintained at 180–200 C to ensure melting of the PS pellets. Since the coupling agents used in this test were all thermoplastic, they may be melted and reformed more than once.

The strings of blended mixture emerging from the TSM were approximately 2.5–5 mm in diameter. Following extrusion, they were chipped into small pellets by a bench top chopper.

The production of transfer-molded wood-fiber/PS composites

Dog bone tensile test samples were produced from the pellets by means of a transfer-molding process. The apparatus used for producing these samples was a Mini-Max Transfer Molder (CS-183 MMX). The nominal sample size of the mold in which the dog bone samples were generated was:

length = 36 mm
width = 16 mm
width (at neck) = 5 mm
thickness = 3 mm

In order to facilitate melting and avoid degradation, the mixture of the wood fiber and PS was kept at a constant 185 C during processing.

Experimental design

A factorial experimental design was carried out to evaluate the mechanical properties of the transfer-molded wood-fiber/PS compos-

TABLE 3. *Moisture content (%) of wood fiber PS composites.*

Fiber loading (%)	Coupling agent type			
	None	PS-PMAA	PS-PVAC	PS-PMAAL
0	0.00[1]			
	(0.00)[2]			
5	0.30	0.17	0.22	0.03
	(0.06)	(0.06)	(0.08)	(0.11)
15	0.43	0.26	0.40	0.24
	(0.11)	(0.10)	(0.13)	(0.11)
25	0.60	0.59	0.43	0.32
	(0.07)	(0.09)	(0.21)	(0.13)

[1] Average values.
[2] Standard deviation.

ites. The two process variables (factors) involved were:

A. Wood-fiber loading levels: 1). 5%, 2). 15% and 3). 25% based on weight.
B. Type of coupling agent incorporated at 1% based on weight: 1). no-coupling agent, 2). polystyrene/poly (methacrylic) acid block copolymer system (PS-PMAA), 3). polystyrene/poly(vinyl acetate) block copolymer (PS-PVAc), and 4). PS-PMAA of low molecular weight (PS-PMAAL).

Specifically, the mechanical properties (response variables) determined were:

1. tensile strength
2. tensile elastic modulus

Testing

Based on this two-way factorial experimental design, an experiment was conducted whereby 5 replicates in each group (total of 65 dog-bone tensile samples) with different combinations of fiber loading and type of coupling agent were manufactured. Tensile tests were carried out on an Instron materials testing machine according to ASTM D638-89 specifications (ASTM 1990). After the tensile test, the moisture content and specific gravity of these samples were measured according to ASTM D 1037-87 (ASTM 1987).

TABLE 4. *Specific gravity of wood fiber PS composites.*

Fiber loading (%)	Coupling agent type			
	None	PS-PMAA	PS-PVAC	PS-PMAAL
0	1.03			
	(0.00)			
5	1.04	1.06	1.05	1.05
	(0.02)	(0.02)	(0.02)	(0.01)
15	1.05	1.05	1.07	1.05
	(0.01)	(0.02)	(0.02)	(0.01)
25	1.06	1.08	1.06	1.07
	(0.02)	(0.02)	(0.02)	(0.01)

TABLE 5. *Tensile elastic modulus (GPa) of wood fiber composites.*

Fiber loading (%)	Coupling agent type			
	None	PS-PMAA	PS-PVAC	PS-PMAAL
0	3.62			
	(0.27)			
5	4.21	4.03	4.07	3.70
	(0.31)	(0.35)	(0.58)	(0.32)
15	4.10	4.03	4.15	3.73
	(0.48)	(0.41)	(0.65)	(0.23)
25	4.09	4.48	4.07	3.84
	(0.69)	(0.56)	(0.32)	(0.34)

RESULTS AND DISCUSSION

Moisture content, specific gravity, tensile strength, and tensile elastic modulus were determined for each combination of coupling agent and polystyrene/wood ratio (Tables 3 through 6). The effect of wood content and type of coupling agent on tensile strength and tensile elastic modulus was analyzed via an analysis of variance (ANOVA) procedure. Since an initial ANOVA indicated that the interaction of coupling agent type and fiber loading was

TABLE 6. *Tensile strength (MPa) of wood fiber PS composites.*

Fiber loading (%)	Coupling agent type			
	None	PS-PMAA	PS-PVAC	PS-PMAAL
0	62.03			
	(2.85)			
5	53.86	59.88	61.04	59.50
	(3.09)	(4.54)	(2.05)	(3.46)
15	51.19	52.68	59.62	58.51
	(3.97)	(3.91)	(1.49)	(1.39)
25	49.00	54.01	56.79	58.29
	(8.37)	(7.64)	(3.22)	(2.39)

TABLE 7. *Two way analysis of variance (ANOVA) by GLM Procedure.*

Source	df	Sum of squares	Mean square	*F* value	Pr > *F*
1. Dependent variable: tensile strength					
Model	5	7.675E 14	1.535E 14	8.44	0.0001
			Type III SS		
Type	3	5.889E 14	1.963E 14	10.79	0.0001
Fiber	2	1.786E 14	8.927E 14	4.91	0.0110
2. Dependent variable: tensile elastic modulus					
Model	5	1.875E 15	3.751E 15	1.89	0.1114
			Type III SS		
Type	3	1.688E 16	5.623E 15	2.83	0.0467
Fiber	2	1.871E 15	9.355E 14	0.47	0.6268

not statistically significant (alpha = 0.05), for both tensile strength and tensile elastic modulus, a second ANOVA was performed excluding the interaction terms. The statistical results are listed in Tables 6 and 7. Throughout the analysis, an overall significance level of 0.05 was chosen.

The moisture content of the samples increased with increasing fiber loading, but to a negligible degree. The specific gravity of the composites ranges from 1.04 to 1.08 and was not significantly correlated with wood-loading percentage.

Tensile strength

Compared to the pure polystyrene samples, for all combinations of fiber loading and type of coupling agent, the average tensile strength decreased (Fig. 1). Both coupling agent type and wood loading levels significantly affected tensile strength (*P* values of 0.0001 and 0.0110, respectively, Table 7). However, this statistical analysis only indicates that at least one fiber/coupling agent combination had a negative effect on the tensile strength as compared to pure PS. Further analysis using the Student-Newman-Keuls (SNK) multiple comparison procedure (Table 8) provides evidence that Groups 2-2, 2-3, and 3-2 have a significant effect. All other combinations of fiber loading and coupling agent had no statistically significant effect on the tensile strength as compared to pure PS. Group 3-2 had a fiber loading of 15% and PS-PMAA as a coupling agent. Groups 2-2 and 2-3 contained no coupling agent and 15% and 25% of wood fiber, respectively. This implies that the reduction in tensile strength by the addition of wood fiber (due to the lack of bonding between the wood and PS) could be compensated by the addition of a coupling agent that functioned to bond the wood fiber to the hydrophobic PS.

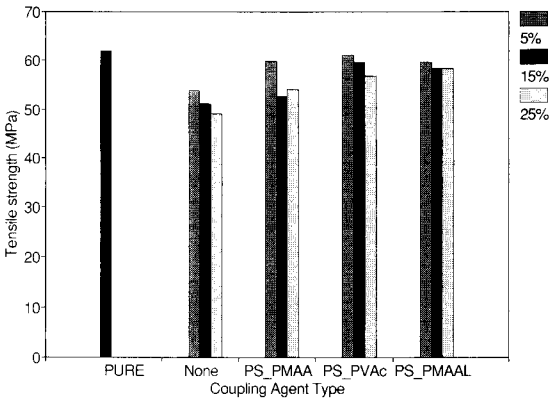

FIG. 1. Tensile strength of wood fiber PS composites compared to pure PS.

Strength relative to type of coupling agent

An analysis of variance indicated that the differences in tensile strength were due primarily to the type of coupling agent being used, as opposed to the selected fiber-loading level (*P* values of 0.0001 and 0.0110, respectively). Type of coupling agent is the factor primarily

TABLE 8. *Multiple comparison using Student-Newman-Keuls (SNK) test by one way ANOVA.*

SNK grouping				Mean[1]	n	Group
1. Dependent variable: tensile strength						
		A		62.03	5	PURE
B		A		61.04	5	PS-PVAc5[2]
B		A	C	59.88	5	PS-PMAA5
B		A	C	59.62	5	PS-PVAc15
B		A	C	59.50	5	PS-PMAAL5
B		A	C	58.51	5	PS-PMAAL15
B		A	C	58.29	5	PS-PMAAL25
B	D	A	C	56.78	5	PS-PVAc25
B	D	A	C	54.01	5	PS-PMAA25
B	D	A	C	53.86	5	None5
B	D		C	52.68	5	PS-PMAA15
	D		C	51.19	5	None15
	D			49.00	5	None25
2. Dependent variable: tensile elastic modulus						
		A		4.48	5	PS-PMAA25
		A		4.21	5	None5
		A		4.15	5	PS-PVAc15
		A		4.10	5	None15
		A		4.09	5	None25
		A		4.07	5	PS-PVAc25
		A		4.07	5	PS-PVAc5
		A		4.03	5	PS-PMAA15
		A		4.03	5	PS-PMAA5
		A		3.84	5	PS-PMAAL25
		A		3.73	5	PS-PMAAL15
		A		3.70	5	PS-PMAAL5
		A		3.62	5	PURE

[1] Means with the same letter are not significantly different.
[2] Type of coupling agent (PS-PVAc) and fiber loading (5%).

responsible for retaining the tensile strength. At a maximum fiber loading of 25%, the most effective coupling agent with respect to countering the deleterious effects of the fibers and retaining the original tensile strength seemed to be PS-PMAAL (Fig. 1).

Strength relative to fiber loading

There is a possibility that a critical fiber-loading level exists. This loading level represents the ideal ratio of fiber to polymer. Increasing the ratio of wood to polymer beyond this level has no further positive physical effects. Reducing the fiber loading to a point below this pivotal ratio could significantly increase the cost of the composite. This concept was conveyed in a further SNK comparison,

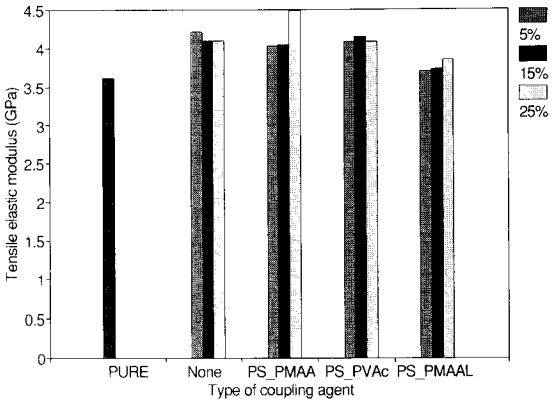

FIG. 2. Tensile elastic modulus of wood fiber PS composites compared to pure PS.

which found that from 15% to 25% fiber loading, the actual fiber content has a negligible bearing upon the tensile strength (Table 8). This is an indicator that a critical fiber-loading level falls between 5% and 15%. It also indicates that more than the critical percentage of fiber may be added without further adverse effects. To do this would, of course, reduce the total cost of the composites, but may present processing difficulties.

Tensile elastic modulus

From the data, a second generalized assumption may be drawn. It can be seen that modulus appears to increase as wood fiber is added to the PS (Fig. 2). This trend appears to be common to all groups, in relation to the average modulus of the pure PS group (Group 1: Table 5). Increases in modulus are apparent from the tabulated results. Samples containing 25% wood fiber and the PS-PMAA type coupling agent exhibited a 23.9% increase. These initial results indicate that adding wood-fiber/coupling agent to the polystyrene creates a composite material that is stiffer than pure PS alone.

The groups that contain PS-PMAA and PS-PMAAL display increased tensile elastic modulus as the fiber loading levels are increased. This would be desirable since the cost of the wood fiber is highly competitive in comparison to most synthetic fibers. At higher fiber

loadings (25%), the most effective coupling agent with respect to increasing tensile elastic modulus was PS-PMAA.

When statistical analysis was employed, however, the differences apparent between the modulus of pure PS and those of the loaded specimens, were not (statistically) significantly different. An analysis of variance (Table 7) conducted on the overall model, where both fiber-loading levels and coupling agent were considered as factors, indicated that the null hypothesis:

modulus of pure PS (grp 1) =
= modulus of loaded samples

cannot be rejected (at 5% significance level). In this case the F test produced a $P = 0.111$.

Tensile elastic modulus relative to type of coupling agent and fiber-loading level

A further ANOVA (combining $SS_{interaction}$ with SS_{error}) also investigated the effect of coupling agent type and fiber-loading level on modulus. The F test for coupling agent type produced a $P = 0.0467$. The same ANOVA confirms that fiber-loading level had a negligible effect on the modulus of the various groups (Table 7).

A further SNK multiple comparison test (Table 8) found no (statistically) significant difference in modulus between the control group (Group 1) and either of the twelve groups, where different coupling agents were used (Groups 2-1–5-3).

Based on this statistical result, it must be concluded that the addition of wood fiber, at any of the 3 selected loadings, or the addition of a coupling agent, did not significantly increase the modulus of the test samples in relation to the modulus of pure polystyrene at significance level = 0.05.

Variability of properties

The statistical evidence seemed to contradict the obvious trends that exist in the data (very large percentage increases in the modulus being apparent in Table 5, ranging from 2.26 to 23.9%, respectively). A feasible explanation for the common finding of statistical non-significance is the high standard deviation that existed within the respective groups of data. This no doubt reflects the fact that there were only 5 replicates per group and data may merely provide an insight into material behavior. It shows that polystyrene containing an additional 5% to 15% TMP aspen fiber displays properties that are not much different from polystyrene itself. Variability in physical properties increased when wood-fiber/coupling agents were introduced into the polystyrene. This variability requires that more samples be tested to verify whether the apparent modulus changes are authentic. It is unknown whether the increased variability is inherent in the material, or due to a processing effects that could be overcome using larger commercial scale processing equipment.

CONCLUSIONS

On the basis of the previous analysis, some conclusions can be drawn:

1. Analysis of variance showed that there was no significant decrease in tensile strength for the groups containing different wood-fiber loading levels when a coupling agent was added. The apparent reduction in tensile strength as the fiber content increases appears to be compensated by the introduction of coupling agent in quantities as low as 1%.
2. At higher fiber loadings (25%), the most effective coupling agent in terms of retaining original tensile strength is PS-PMAAL.
3. At higher fiber loadings (25%), the most effective coupling agent for increasing tensile elastic modulus is PS-PMAA.
4. To confirm this tabulated result, a further test using a larger sample size is required.
5. Increased fiber loading (more than 25%) is suggested, and a further investigation is needed to confirm both cost and performance effectiveness of the composites.

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REFERENCES

AMERICAN SOCIETY FOR TESTING AND MATERIALS. 1987. Standard method of evaluating the properties of wood-based fiber and particle panel materials. ANSI/ASTM D 1037-87. Philadelphia, PA.

———. 1990. Standard method for tensile properties of plastics. Vol. 08.01. Plastics (1):D 638-89. Philadelphia, PA.

BIRD, R. B., R. C. ARMSTRONG, AND O. HASSANGER. 1987. Dynamics of polymeric liquids, 2nd ed. Vol. 1. John Wiley and Sons, New York, NY.

BRANDUP, J., AND E. H. IMMERGUT. 1989. Polymer handbook, 3rd ed. John Wiley and Sons, New York, NY.

HON, D. N.-S., W. Y. CHAO, AND C. J. BUHION. 1992. Commingled plastics/newspaper composites. Plastics Eng. 48(10):25–28.

MALDAS, D., AND B. V. KOKTA. 1989. Improving adhesion of wood fiber with polystyrene by the chemical treatment of fiber with a coupling agent and the influence on the mechanical properties of composites. J. Adhes. Sci. Technol. 3(7):529–539.

———, AND ———. 1990a. Effects of coating treatments on the mechanical behavior of wood fiber-filled polystyrene composites. I. Use of inorganic salt/polyvinyl chloride and isocyanate as coating components. J. Appl. Polymer Sci. 40:917–928.

———, AND ———. 1990b. Effects of coating treatments on the mechanical behavior of wood fiber-filled polystyrene composites. II. Use of inorganic salt/polyvinyl chloride and isocyanate as coating components. J. Reinf. Plastics Compos. 8:2–12.

———, AND ———. 1990c. Influence of phthalic anhydride as a coupling agent on the mechanical behavior of wood fiber-polystyrene composites. J. Appl. Polymer Sci. 41:185–194.

———, AND ———. 1991. Role of coupling agents and treatments on the performance of wood fiber-thermoplastic composites. Poster #12. 1991 Woodfiber-Plastic Composites Conference, April 28–30, Madison, WI. The University-Industry Research Program, University of Wisconsin and Wisconsin–Madison, and the USDA-Forest Service, FPL.

MEIR, D. J. 1987. Theoretical aspects of block copolymers. Pages 269–301 *in* N. R. Legge, G. Holden, and H. E. Schroeder, eds. Thermoplastic elastomers—A comprehensive review. Hanser Publishers, Munich, Germany.

SEAN, SY. T., B. SANSCHAGRIN, B. V. KOKTA, AND D. MALDAS. 1990. Effect of treated wood fiber-polystyrene composites under prolonged immersion in water. Mokuzai Gakkaishi. 36(8):637–643.

———, ———, ———, AND ———. 1991. Effect of processing variables on the mechanical properties of composites of compression molded polystyrene-wood fiber composites. Die Angewandte Makromolekulare Chemie 184: 157–166.

U.S. CONGRESS, OFFICE OF TECHNOLOGY ASSESSMENT. 1989. Facing America's trash: What next for municipal solid waste. OTA-O-424. U.S. Government Printing Office, Washington, DC.

VAN KREVELEN, D. W., AND P. J. HOFTYZER. 1976. Properties of polymers. Elsevier Scientific Publishing Co., Amsterdam, Netherlands.