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# WOOD-DERIVED PRODUCTS IN CHEMICALLY RECYCLED POLYSTYRENE (PS) LITERATURE REVIEW

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Polystyrene (PS) is one of the most produced thermoplastics in the world, which has been widely used in packaging, e.g., expanded foams and containers, and electronics. Addition of wood-derived products, including cellulose fibres and lignin, into PS matrix as fillers, reinforcement, and functionalities, has been studied and reported. With the impact of plastics, especially packaging materials, on the environment, more and more attention has been paid to reuse, reduction, and recycling of such materials. An important characteristic of PS is that it can be chemically recycled: a process where PS is depolymerized to the original monomer via pyrolysis. After purification, these monomers can be reused to produce PS. Chemical recycling PS has been commercialized, and there are plants in Canada, including Polystyvert and Pyrowave. This report reviews the current status of PS chemical recycling, the application of wood-derived products in PS, and the potential effect of such additives in PS chemical recycling. The purpose of this report is to provide insights of potential opportunities in this field for FPInnovations.

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# 1 INTRODUCTION OF POLYSTYRENE

## 1.1 Polystyrene Structure and Properties

Polystyrene (PS) is a synthetic polymer made from petroleum-based products. PS is polymerized via free radical polymerization primarily by bulk and suspension processes. The molecular structure and the polymerization process of PS is shown in Figure 1.

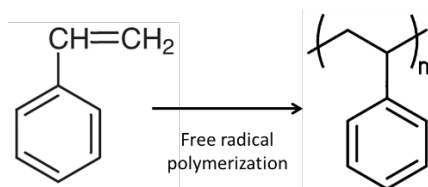


Figure 1. Polymerization process of polystyrene.

Depending on the distribution of the phenyl groups on the two sides of polymer chains, PS has three types of tacticity: atactic, syndiotactic, and isotactic. Of these three types, the most produced one is the atactic form, where the phenyl groups are randomly distributed. Owing to the structure, this type of PS does not crystallize, which impart PS high transparency. Commercial production of syndiotactic PS (SPS) was realized by Idemitsu corporation with higher price compared to the regular PS. According to the company, SPS has better heat resistance, electrical properties, and chemical resistance than atactic PS.<sup>1</sup> Styrene is also widely used by copolymerizing with other monomers to form different materials, such as acrylonitrile butadiene styrene (ABS) – an engineering plastic – and styrene-butadiene rubber (SBR) – an elastomer. Isotactic polystyrene is not produced in large scale. In this report, the focus is on atactic PS, which is referred as PS.

Table 1. Typical values of some PS properties.<sup>2</sup>

Property	Unit	Value
Density	g/cm <sup>3</sup>	1.07
Surface Hardness	-	RM80
Tensile Strength	MPa	34
Flexural Modulus	GPa	3
Notched Izod	kJ/m	0.02
Elongation at Break	%	1.6
Strain at Yield	%	1.4
Max. Operating Temperature	°C	50
Water Absorption	%	0.05
Volume Resistivity	log ohm.cm	16
Dielectric Constant 1 kHz	-	2.6
HDT @ 0.45 MPa	°C	90
HDT @ 1.80 MPa	°C	80
Melting Temp. Range	°C	200 - 250
Mould Shrinkage	%	0.5
Mould Temp. Range	°C	20 - 50



PS has glass transition temperature of  $107^{\circ}\text{C}$ ,<sup>3</sup> thus it is a rigid material at room temperature. The values of some PS properties are listed in Table 1. General PS is brittle, which limits the application. By incorporation of soft segments (e.g., polybutadiene) into PS, high impact polystyrene, or HIPS, can be produced.



Figure 1. Examples of XPS (Styrofoam, left) and EPS (right).

PS can also be processed to foam via extrusion or molding, which are called extruded polystyrene (XPS) foam and expanded polystyrene (EPS) foam, respectively. There has been misapplication of the name Styrofoam for all PS foams, which is a trade name of the Dow Chemical Company specifically for XPS. The appearance of these two types of PS foams is shown in Figure 1. Both XPS and EPS need to use blowing agents during processing to create the bubbles in the foams. By controlling the loading of blowing agents, the properties of foams can be tuned. Some properties of EPS with different densities are shown in Table 2.

Table 2. Some physical and mechanical properties of ESP.<sup>4</sup>

Physical Property	Standard	Unit	Results		
Apparent minimum density	DIN 53420	$\text{Kg/m}^3$	10	20	30
Thermal conductivity (at $-10^{\circ}\text{C}$ )	DIN 52612	$\text{W/m}^{\circ}\text{C}$	0.036-0.038	0.033-0.036	0.031-0.035
Tension compression with 10% deformity	DIN 53421	$\text{Kg/cm}^2$	0.6–1.1	1.1–1.6	2.0–2.5
Permanent resistance to compression with <2% deformity		$\text{Kg/cm}^2$	0.15–0.25	0.25–0.40	0.45–0.60
Flexibility resistance	DIN 53423	$\text{Kg/cm}^2$	0.6-3.0	1.50–3.9	3.3–5.7
Elasticity modulus (compression tests)	DIN 53457	$\text{Kg/cm}^2$	16-52	34-70	77–113
Absorption of immersing water (7 days)	DIN 4108	%	0.5-1.5	0.5-1.5	0.5-1.5
Permeability to water vapor	DIN 52615	$\text{g/m}^2\text{d}$	40	35	20

The wide application of PS has caused health and environmental concerns. Since PS is not a biodegradable plastic, the disposed PS products remained in the environment for a long time and slowly fracture to small particles, which are typically called microplastics. These particles may interact with other contaminants in the environment, such as heavy metals, organic matter, and microorganisms, and migrate to the food chain eventually.<sup>5</sup> In addition to that, the foam form PS is difficult to recycle due to its bulky volume.<sup>6</sup>





## 1.2 Polystyrene Applications and Market

The applications of PS can be categorized based on its two forms: rigid PS and PS foam. The rigid PS can be found in many consumer products, including cutlery and dinnerware, appliances, automotive parts, electronic device housings, toys, as well as laboratory contains, such as Petri dishes, test tubes, and microplates. Some of the examples regarding rigid PS applications are given in Figure 2. PS foams contain more than 95% air and have good thermal insulation and damping properties. Therefore, they are broadly used in packaging, food containers/trays, and insulation board in construction.<sup>7</sup>



Figure 2. Examples of rigid PS applications.

PS is one of the most produced thermoplastics in the world with production of 26.6 million metric tons in 2020.<sup>8</sup> The market value of PS was USD 9.5 billion in 2021 and expected to increase to USD 10 billion by 2028, driven by the increasing demand from the consumer electronics industry.<sup>9</sup> PS is produced worldwide by many companies, including Trinseo, BASF, Alpek, Sabic, Formosa Chemicals & Fibre, Total Energies, Kumho Petrochemical, INEOS, etc.

Even though PS has been widely used, it is not effectively recycled, especially PS foams, owing to the bulky nature of the materials. This has caused increasing concerns regarding environment protection, because the PS foams occupy a large volume of landfill space even though they are a small component by weight. One of the approaches to recycle PS is chemical recycling, which has become the trend for PS industry.<sup>9</sup> More information about chemical recycling PS will be introduced in Section 3.



## 2 POLYSTYRENE COMPOSITES WITH WOOD DERIVATIVES

Overall, PS is most applied as transparent films/sheets and foams (EPS), and the research about PS-based composites is not as intense as for other plastics, such as polypropylene (PP). In this section, the focus will be on PS-based composites filled with wood derivatives, although other fillers have been reported as well, such as silica,<sup>10</sup> clay,<sup>10,11</sup> aluminium,<sup>11</sup> and carbon nanotubes.<sup>12</sup> The PS composites with natural fibres other than the wood-sourced ones will also be reviewed, because such fibres have similar composition and processing approaches as the wood fibres.

### 2.1 Polystyrene Composites with Cellulose Materials

Since cellulose is a polar hydrophilic material, which is not compatible with nonpolar hydrophobic PS, surface modification and/or dispersing/coupling agents are typically required to improve the dispersion of cellulose materials as well as to improve the interactions at the interface to achieve better performance. One of the research interests in PS-based composites is the application of cellulose nanomaterials, including both cellulose nanocrystals (CNC) and cellulose nanofibrils (CNF). Owing to the strong hydrogen bonding interactions between cellulose nanomaterials, dispersion of such materials is typically the major challenge. Different approaches have been reported to improve the dispersion. For example, the cellulose surface can be modified with small molecules or grafting with polymer chains. Such modified nanomaterials can be mixed with PS by melt processing. On the other hand, uniform dispersion can be achieved by using proper processing technique, including organic solvents media and emulsion template. In the solvent method, both cellulose nanomaterials and PS are dissolved in an organic solvent, e.g., dimethylformamide (DMF), followed by casting or electrospinning. For the emulsion method, the styrene is typically mixed with water to form Pickering emulsions stabilized with cellulose nanomaterials, and PS nanocomposites are obtained by polymerizing the monomer. Owing to the good dispersion of cellulose nanoparticles or fibre in PS, such nanocomposites typically possess high transparency similar to PS. The appearance of such PS nanocomposite films and a schematic illustration of the emulsion template approach are shown in Figure 3. The literature regarding PS/CNC and PS/CNF nanocomposites is summarised in Table 3.

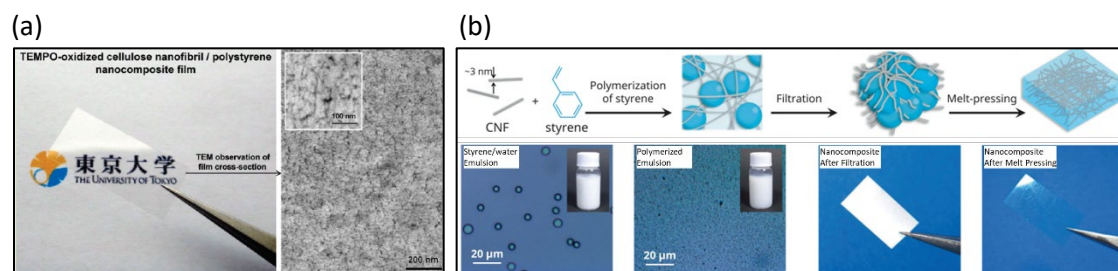


Figure 3. (a) CNF/PS nanocomposite film prepared by casting from DMF, the appearance and TEM images,<sup>13</sup> and (b) illustration of the emulsion template method to prepare CNF/PS nanocomposite.<sup>14</sup>





Table 3. Summary of literature regarding PS based nanocomposites with cellulose nanomaterials: CNC and CNF.

Filler	Modification/ Compatibilization	Processing Method	Functions/Properties	Ref.
CNC	Surface modification with benzoic acid anhydride in THF	Freeze drying CNC with PS powder in water → Extrusion at 200 °C → injection molding	<ul style="list-style-type: none"> <li>The addition of 5 wt% CNCs to the PS matrix resulted in a 30% improvement in tensile strength and a 23% increase in Young's modulus</li> <li>The improvement of properties was attributed to the intense entanglement of PS chains and the modified CNCs due to <math>\pi</math>-<math>\pi</math> interactions between benzene rings</li> </ul>	15
	Surface grafting with short chain PEG → adsorption of long chain PEO	Freeze drying CNC/PEO mixture in water → Extrusion at 200 °C	<ul style="list-style-type: none"> <li>Presence of PEG on CNC surface improved adsorption of PEO, avoiding thermal degradation of CNC during PS processing. Thus, the composite films was less dark than the one without PEG grafting</li> <li>The CNC-PEG-PEO/PS composites showed improved dynamic properties above T<sub>g</sub> and water vapor barrier performance</li> </ul>	16
	None	Dissolving CNC and PS in an organic solvent mixture (DMF/THF) → electrospinning	<ul style="list-style-type: none"> <li>PS and CNCs were physically mixed and CNCs were probably distributed on the surface of the electrospun nanofibers</li> <li>Thermal properties of electrospun PS/CNC nanocomposites were enhanced by increasing CNC loading levels</li> <li>The tensile strength of electrospun PS/CNC nanofibrous mats was elevated 170% by incorporating 7 wt% CNCs in PS matrix, while the ductility was decreased from 60 to 25%</li> <li>The hydrophobic property could be increased to 138° when 1 wt% CNCs was incorporated</li> </ul>	17
	None	Dissolving CNC and PS in an organic solvent mixture (DMF/THF) → electrospinning	<ul style="list-style-type: none"> <li>The CNC loading was 1 to 10 % and the presence of CNC has none or negative effect on the mechanical properties of the spun mat</li> <li>The orientation of the fibres played more important role in determining the mat properties</li> </ul>	18
	None	Formation of CNC stabilized styrene-in-water emulsion followed by polymerization	<ul style="list-style-type: none"> <li>During polymerization, the emulsion was either stirred mechanically or treated with ultrasonication</li> <li>Ultrasonication treatment accelerates polymerization and lower the reaction temperature compared to the mechanically stirred one: to reach 85% styrene conversion, the reaction time was reduced from 3 h to 15 min, the reaction temperature was decreased from 75 °C to 45 °C</li> </ul>	19



Filler	Modification/ Compatibilization	Processing Method	Functions/Properties	Ref.
CNF	TEMPO oxidization (TOCN)	Dissolving PS in DMF and mix CNF in it → casting in glass petri dishes	<ul style="list-style-type: none"> <li>• TOCN/PS nanocomposite films exhibited high optical transparencies</li> <li>• The tensile strengths, elastic moduli, and thermal dimensional stabilities were increased</li> </ul>	13
	None	Formation of styrene-in-water Pickering emulsion stabilized with CNF → polymerization → filtration	<ul style="list-style-type: none"> <li>• CNF/PS films were prepared through the emulsion template method, where the films are composed of particles with structures of PS as the core and CNF as the shell</li> <li>• Such films can be used for biomedical or cosmetic applications, owing to the functionality of CNF on the particle surfaces</li> <li>• Transparent films with improved mechanical properties can be prepared via melt-pressing</li> </ul>	14
	As is or treated with deep eutectic solvents	Dissolving PS in dichloromethane (DCM) → formation of PS-in-water Pickering stabilized with CNF → evaporation of liquid → hot pressing	<ul style="list-style-type: none"> <li>• Final mechanical performances of the composites are largely determined by the Pickering emulsion morphology</li> <li>• The emulsification ability of CNF depended on the O/W ratio, content, fiber diameter and surface hydrophilicity of CNF</li> <li>• When the additional amount of cellulose was 8 wt. %, the emulsion droplets formed by CNF was smaller and more uniform. In particular, the 8 wt. % of treated CNF showed remarkable enhancement in mechanical properties of PS composite</li> </ul>	20
	Phenyl-isocyanate	Mixing modified CNF and PS in DMF → coagulate the solution in methanol to get composite beads → hot pressing	<ul style="list-style-type: none"> <li>• The elastic modulus of the PS-iCNF composite increased gradually, while the fracture bending strength decreased as the loading increased</li> <li>• Failure behavior of the composite changed from ductile to brittle fracture with increasing iCNF loading</li> <li>• The effect was ascribed to phenyl isocyanate, which generates <math>\pi - \pi</math> interactions with the phenyl group on the polymer chains</li> </ul>	21



PS composites with wood fibres were also reported, including both rigid PS<sup>22–25</sup> and EPS<sup>26–28</sup> and a summary of the literature is given in Table 4. For the composites with rigid PS, addition of wood fibre was mainly to improve the mechanical properties of the materials. For such applications, a coupling agent is typically critical for the performance of the composites. Several types of coupling agents have been reported, including methylene diphenyl isocyanate (MDI), silanes, and copolymers of styrene and maleic anhydride. The main purpose of coupling agents is to improve the interface between cellulose fibre and PS polymer, as well as facilitate the dispersion of fibre. Coupling agents carrying phenyl rings are preferred because they can impart  $\pi$ - $\pi$  interactions with the phenyl rings on PS polymer chains. An example of the working mechanisms of coupling agents is given in Figure 4. This type of composites was typically prepared via melt processing, *e.g.*, extrusion and compounding in an internal mixer.

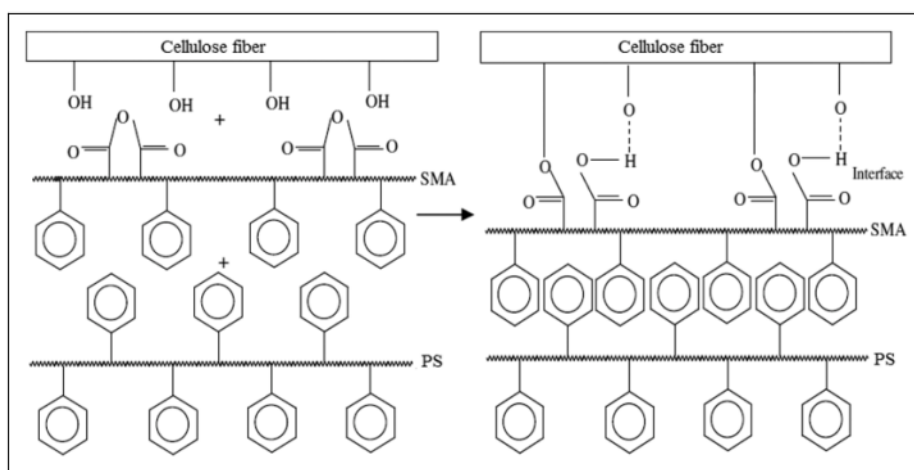


Figure 4. Schematics of the interactions between cellulose fibre and PS with the presence of poly(styrene-co-maleic anhydride) oligomer (SMA) coupling agent.<sup>24</sup>

Composites of EPS with cellulose fibres were studied as well. For this type of material, recycled EPS foams were normally used from the environmental and economical perspective. There are two applications in literature for such composite: gypsum board with lower density and better thermal insulation for construction and passive radiative cooling materials.

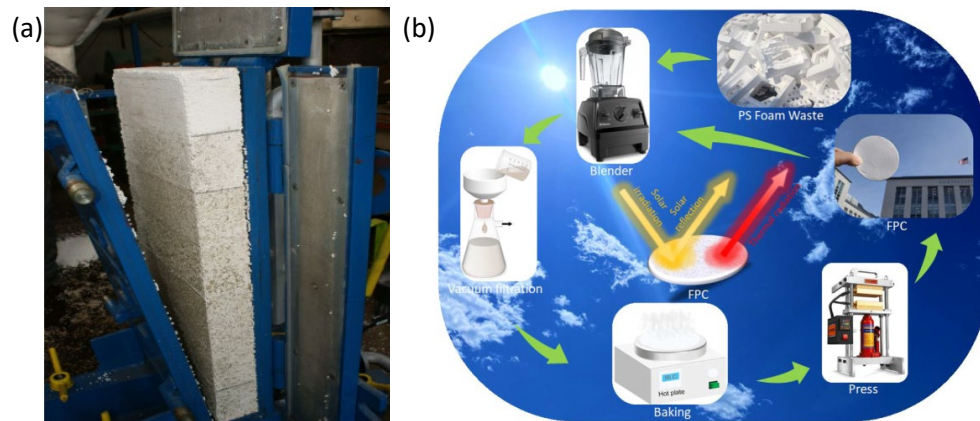


Figure 5. (a) Hybrid thermal insulation material consisting of EPS, recycled cellulose fibre, binder, and assorted additives; (b) Schematic illustration of the closed-loop fabrication and recycling process of foam-paper composite (FPC) and its radiative cooling mechanism.<sup>28</sup>



Table 4. Summary of literature regarding PS based composites with wood fibres.

Filler	Modification/ Compatibilization	Processing Method	Functions/Properties	Ref.
<b>Bleached pine fibres</b>	MDI or blocked MDI was used as coupling agent	PS was mixed with fibres in an internal mixer with/without coupling agents	<ul style="list-style-type: none"> <li>The mechanical properties, including both tensile and flexural properties, were improved with the addition of 30% pine fibres</li> <li>Application of MDI or phenyl blocked MDI can further improve the performance of composites</li> <li>The improvement was ascribed to the covalent bonds between MDI and the surface hydroxyl groups on fibres, as well as the <math>\pi - \pi</math> interactions between the phenyl rings on MDI and PS</li> </ul>	22
<b>Unbleached and bleached CTMP from aspen, TEMBEC 6816</b>	Coupling agents: Poly[methylene(polyphenyl isocyanate) (PMPPIC), Vinyltri(2-Methoxyethoxy) silane (A-172), and Gamma-Methacryloxypropyltrimethoxy silane (A-1100)	Treated pulp was mixed with PS in roll mill at 145-225°C → grinding → hot pressing	<ul style="list-style-type: none"> <li>The preferred processing temperature is 175 °C</li> <li>The tested fibre contents were 10, 20, 30, and 40% in composites</li> <li>Coupling agents are important in improving mechanical properties and the effect of different coupling agents were compared</li> <li>Coating followed by an isocyanate treatment is the most effective</li> <li>Silane treatment is inferior to other treatments</li> <li>Covalently linked polystyrene to cellulose in grafted fiber provide a superior interfacial area</li> </ul>	23
<b>Bleached softwood pulp fibre</b>	Ground the pulp to length between 100 µm and 270 µm; poly(styrene-co-maleic anhydride) oligomer (SMA) as coupling agent	Extrusion through a twin-screw extruder	<ul style="list-style-type: none"> <li>Coupling agent is critical to improve the interfaces between fibres and PS matrix for better performance</li> <li>The mechanical properties were substantially improved with the application of coupling agent</li> <li>High cellulose fiber loading (more than 20 wt%) caused decrease in mechanical properties</li> <li>Three extrusion speed were compared: 200 rpm, 400 rpm, and 600 rpm</li> <li>Composites processed at 400 rpm presented higher mechanical properties and better dispersion of the fibres</li> </ul>	24,25
<b>CTMP</b>	Graft PS on pulp fibre surfaces via polymerization	Mixing in an internal mixer → injection or compression molding	<ul style="list-style-type: none"> <li>Grafting polymers on fibre surfaces improves mechanical properties of composites</li> <li>The injection molded samples had better performance than the one prepared by compression molding</li> </ul>	29



Filler	Modification/ Compatibilization	Processing Method	Functions/Properties	Ref.
<b>Bleached wood fibre</b>	None	Mixing gypsum, crushed EPS, and fibres in water and dry in a mold for 6 days	<ul style="list-style-type: none"> <li>Recycled EPS and gypsum were used</li> <li>Lower density than pure gypsum board</li> <li>Better heat insulation than gypsum board</li> <li>Lower flexural and compression strength, but meet minimum strength requirement of EN 13279-1</li> </ul>	26
<b>Recycled cellulose fibres</b>	None	Mixing fibres with pre-expanded PS, interphase material (cement or gypsum), flame retardant, fungicide, and binder followed by drying in a mold	<ul style="list-style-type: none"> <li>Hybrid thermal insulation material was prepared</li> <li>Expanding graphite was used as flame retardant</li> <li>Selection of materials, compositions, mixing sequence were studied</li> <li>Fibers with length of less than 1mm had the most preferred set of features, allowing for their even distribution at the interfaces on the EPS granules</li> </ul>	27
<b>Print paper</b>	None	Blend PS waste foam with print paper in a high-speed blender → filtration + drying → pressing	<ul style="list-style-type: none"> <li>The obtained foam-paper composite (FPC) is a passive radiative cooling material</li> <li>The FPC has 96% solar reflectance and 80% emittance in the infrared wavelength range</li> <li>Theoretical demonstration showed that the FPC can achieve continuous daytime radiative cooling with an average temperature drop of 6 °C under direct sunlight</li> </ul>	28





## 2.2 Polystyrene Composites with Lignin

Lignin is the second-most abundant component of plant material only after cellulose. It has been proven to be a promising natural resource in various applications, for example, adhesives and coatings, foam, carbon fiber, light-colored UV absorber, antioxidants, etc.

Lignin is a promising compound to be used in polymers because of its phenolic structure which could lead to improvement of mechanical properties, thermal stability, photodegradation strength and UV absorption. However, the influence of lignin on thermomechanical and physical properties of synthetic polymers depends on the degree of miscibility between lignin and the polymer. In terms of lignin application in recycled PS, a preliminary literature survey indicated that only a few research studies have been done in this area. PS is not very compatible with other polymers and materials. In addition, the polar nature of lignin increases its immiscibility with PS. Therefore, the formation of PS-lignin with adequate miscibility is challenging. Modifying chemical structure of lignin could be a potential way to improve the compatibility between lignin and PS as the chemical modification of lignin could increase its hydrophobicity, and consequently facilitate dispersion in organic solvents. In addition, the increase in the carbon aliphatic chain performed via replacing hydroxyl groups per ester groups on lignin could reduce its polarity and increase its solubility in nonpolar solvents. The modification by introducing double bonds with maleic anhydride or methacrylic could also cause a reduction in glass transition temperature of the lignin, which significantly increases its solubility in organic solvents. This allows modification of the mechanical properties when lignin is dispersed in polymeric matrices.

PS-lignin composites from recycled PS with maleated lignins have been studied and produced through a reactive extrusion process<sup>30</sup>. In this study, lignin was modified by a reaction with maleic anhydride. The maleated lignin was then incorporated to recycled PS (rPS) at 2, 5, and 10 wt% by a melt-blending process. In these processes, maleic anhydride can esterify the hydroxyl groups of lignin to maleate and form a half-ester with a double bond, which can further react with nonpolar PS in a mechanism shown in Figure 6. The study revealed that both rPS and rPS/maleated lignin composites had similar T<sub>g</sub> values and the decomposition profiles in only one step, suggesting some degree of miscibility between modified lignin and the recycled PS polymer. The thermal stability of rPS with 2 and 5% of maleated lignin was slightly greater than that of the virgin PS, but a decrease of thermal stability of rPS with 10% of modified lignin content was observed.

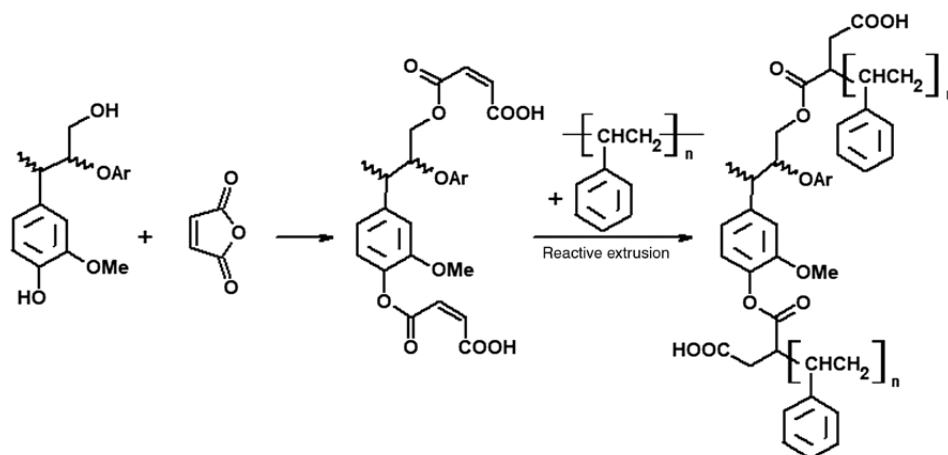


Figure 6. Esterification reaction of lignin with maleic anhydride and proposed grafting reaction with polystyrene.<sup>30</sup>

Pérez-Guerrero et al have studied the morphology and thermo-mechanical properties of recycled PS-based materials (rPS) reinforced with 5% and 15% lignin from kraft pulping of *Eucalyptus nitens*. In this study, the kraft lignin was modified through two esterification processes using either acetic or maleic anhydride. The lignin was then incorporated into rPS by melt mixing. Torque variation in the process was evaluated and compared to those of neat rPS and virgin PS. The results showed a plasticizing effect of lignin with a reduction in the torque values. The morphology by SEM showed a particle size reduction of acetylated and maleated lignin and a more homogeneous dispersion at 5%. In the thermal behavior of the composites, DSC did not show significant changes in the glass-transition temperature of rPS with the presence of lignin, whereas TGA indicated that the thermal stability was improved for composites containing 5% esterified lignin. An increasing storage modulus with increasing lignin content was observed by dynamic mechanical analysis (DMA). The flexural modulus also increased with lignin addition. According to the results showed in the TGA, DMA and flexion tests, it was suggested that the best performance was observed on rPS with 5% esterified lignin, but using 15% resulted in a reduction of the thermal and mechanical properties. Acetylated lignin had better performance than the maleated lignin<sup>31</sup>.

Zhang et al reported lignin-PS composite foams, where kraft lignin was first modified to butyrate lignin by esterification in order to increase the compatibility as fillers with both monomer and polymer. The modified lignin was then used to make composites with PS through bulk polymerization and high internal phase emulsion (HIPE) polymerization. It was found that the incorporation of modified lignin in the composites could increase the Young's modulus significantly and almost double (1,391 MPa) at 15 wt% of lignin loading as compared with pure PS foam, and excellent porous structure and mechanical properties were maintained with the lignin content as high as 10 wt% of the total foam mass<sup>32</sup>.

Schorr et al have also studied recycled PS-lignin composites by adding unmodified and maleated kraft lignins into recycled PS as fillers at different concentrations (0, 10, 20 and 30%) via melt blending<sup>33</sup>. The recycled PS composites with 20% of maleated Kraft lignins were also prepared by applying a previous surface treatment to the maleated Kraft lignins with recycled PS in solution. A complete mechanical, thermal and morphological characterization was performed on the recycled PS-based composites containing kraft lignins. In the morphological study, they found better compatibility with recycled PS for unmodified kraft lignins than maleated kraft lignins except for the composite where maleated lignins



surface treatment was applied. In this case, the recycled PS was well incorporated in the porous structure of maleated kraft lignins. Their results also showed that addition of unmodified kraft lignins led to better mechanical and thermal properties of the composites than when maleated Kraft lignins were added. For the composite formulated with 20% maleated kraft lignins with a previous surface treatment, the properties were improved to become comparable to those of composites with unmodified kraft lignins. The poor mechanical and thermal properties of recycled PS composites with maleated kraft lignins could be related to the porous structure of the maleated kraft lignins.

A polymeric composite was also formed by the combination of natural or esterified lignin and styrene through a sequential mass-suspension polymerization process<sup>34</sup>. In this study, kraft lignin from eucalyptus wood was esterified with methacrylic anhydride to ensure organic phase homogeneity into the reaction medium. Comparatively, the esterified lignin displayed an increase of its glass transition temperature, and the styrene/lignin-based polymers exhibited higher average molar masses in comparison to the values observed for polystyrene synthesized with similar amounts of benzoyl peroxide, due to the ability of lignin to act as a free-radical scavenger. The synthesized composites presented regular morphology and proper lignin dispersion. The authors claimed that based on the very simple polymerization system, it is possible to enhance the final properties of polystyrene through the incorporation of lignin, providing an important platform for developing attractive polymeric materials from renewable resources. No performance properties have been specified for comparison between the PS composites with and without the incorporation of native or esterified lignins in this report.

In summary, PS is not very compatible with lignin due to the difference of polarity which increases immiscibility between lignin and the PS polymer. A preliminary literature survey indicated that a few studies have been done in incorporating lignins with recycled PS. Some studies indicated that chemically modified lignins by esterification could increase the compatibility between lignin and PS to form lignin-PS composites with the lignin addition as high as 10 wt%, leading to improved performance in mechanical and thermal properties. However, higher lignin additions showed contrary behavior. More work needs to be done and the latest research progress in this area is to be followed to understand the impacts of the technical lignin types and chemical modification methods on compatibility between lignins and PS, and, as a result, on the thermal stability and mechanical properties of resulting PS-lignin composites.

## 2.3 Polystyrene Composites with Other Wood Derivatives and Natural Fibres

In addition to the wood derivatives introduced in sections **Error! Reference source not found.** and **Error! Reference source not found.**, other materials, such as sawdust,<sup>35–37</sup> wood flour,<sup>38–42</sup> primary sludge,<sup>43</sup> and wood chips,<sup>44</sup> were also mixed with PS for composite applications. A summary of such applications is given in Table 5. Generally, many of these works aim to utilize PS waste, especially EPS, for composite, where performance is not the key objective. Thus, coupling agents were not necessary. In order to reduce the bulky volume of EPS, organic solvents, *e.g.*, acetone and gasoline, were used to dissolve EPS in some cases, which is a concern for safety during processing and may have negative environmental impact. There was also research investigating to replace existing foaming agent with high pressure CO<sub>2</sub>.



However, the high requirements for the processing conditions may hinder the application of this method.

PS composites with natural fibres were also reported in literature<sup>45–48</sup> and the summary is listed in Table 5 as well. Since natural fibres mainly consist of lignocellulose, the compatibilization and processing are similar to the ones containing wood derivatives. The research interest is the improvement of dispersion of fibres and the interface between fibres and PS. Utilization of recycled PS foams as the matrix material was also reported.



Table 5. Summary of literature regarding PS based composites with other wood derivatives and natural fibre.

Filler	Modification/ Compatibilization	Processing Method	Functions/Properties	Ref.
<b>Sawdust</b>	None	Dissolving EPS in solvent → drying → grinding → mixing with sawdust → compression molding	<ul style="list-style-type: none"> <li>Recycled EPS was used</li> <li>The EPS was dissolved in a solvent to reduce the volume</li> <li>The composite containing 50% of sawdust demonstrated mechanical properties comparable to a commercial plywood</li> <li>Composites filled with rice husk were prepared using the same method as well</li> <li>The properties of PS/sawdust composites can be further improved by addition of carbon additives recycled from vehicle soot</li> </ul>	36,37
<b>Sawdust</b>	None	Dissolving EPS in gasoline at high solid content → mixing with filler by kneading	<ul style="list-style-type: none"> <li>Waste PS was mixed with gasoline at high concentration, making a dough-like material, add fillers were added by kneading</li> <li>The gasoline was also used as a foaming agent during processing</li> <li>Three different fillers were compared: wheat flour, wheat starch, sawdust of MDF. The one loaded with sawdust had the best performance</li> </ul>	35
<b>Wood flour</b>	None	Mixing wood flour with PS in an internal mixer at 185 °C → granulation → hot pressing at 230 °C	<ul style="list-style-type: none"> <li>Composite samples were saturated with carbon dioxide at high pressure and ambient temperature and the saturated specimens were expanded at warm glycerin bath to form EPS</li> <li>The EPS composites contained 10 % and 20 % wood flour</li> <li>Impact and tensile properties of EPS composites and foaming process and structure were studied</li> <li>Addition of wood flour improved mechanical properties of EPS composites</li> <li>Fiber content was the most important parameter controlling impact strength and tensile modulus</li> </ul>	40,41
<b>Wood flour</b>	Maleic anhydride (MA)	Mixing wood flour with PS in an internal mixer at 235 °C	<ul style="list-style-type: none"> <li>Composites contained 35, 45, and 55% wood flour were prepared</li> <li>MA was used as coupling agent for all polymer matrices (may not effective), UV-stabilizer and paraffin wax were added</li> <li>The composites possess improved flexural modulus, but decreased flexural strength and tensile properties</li> </ul>	42



Filler	Modification/ Compatibilization	Processing Method	Functions/Properties	Ref.
<b>Wood flour</b>	Struktol TR065, a commercial compatibilizer	Compounding in an internal mixer	<ul style="list-style-type: none"> <li>Both virgin PS and recycled EPS were used. The recycled EPS was pressed at 120 °C to reduce the volume</li> <li>Addition of 50% of wood flour increased the flexural modulus of both recycled and virgin PS</li> <li>Tensile modulus showed decreasing trend with higher wood contents</li> <li>The best Izod impact strength was from the recycled PS composite containing 30% wood flour</li> <li>At 2% loading of the selected compatibilizer, there was no effect on the performance of composites</li> </ul>	38
<b>Wood particles (5-10 mm)</b>	None	Mixing EPS beads, urea formaldehyde resin, and fire-retardant agent → foaming in a mold	<ul style="list-style-type: none"> <li>Lightweight composites were prepared using EPS and wood particles</li> <li>The fire resistance was improved by addition of melamine phosphate (MPP) and aluminum hypophosphite (AHP)</li> </ul>	39
<b>Primary sludge &amp; fly ash</b>	PS-g-MA	Mixing PS, PS/PS-g-MA/primary sludge/fly ash → foaming	<ul style="list-style-type: none"> <li>PS/PS-g-MA/primary sludge/fly ash were mixed at different ratios</li> <li>Chemical reactions occurred between PS, primary sludge, and fly ash</li> <li>Primary sludge and fly ash were uniformly dispersed into PS, increasing the mechanical properties and decreasing the melting point of the PS foam composites</li> <li>The sound-absorbing composite produced had qualified ISO 11654:1997 on the rating level of the sound absorption coefficient on materials for rooms with sound absorption classes D and C with the value of <math>\alpha_w</math> 0.328-0.793</li> </ul>	43
<b>Wood waste from chips (sieved with a 5mm mesh)</b>	None	Dissolving EPS waste in acetone → Mixing with wood chips via extrusion → drying	<ul style="list-style-type: none"> <li>Recycled EPS was used</li> <li>The EPS was dissolved in acetone to reduce the volume</li> <li>The open porosity and the absorption of water by the samples decrease as the binder content increases</li> <li>For the same binder content, thermoforming reduces porosity by around 25% while improving the internal structure of the samples</li> </ul>	44
<b>coconut husk fiber (coir) and banana stem fiber (BSF)</b>	NaOH solution	Cast in organic solvent (mixture of acetone and thinner)	<ul style="list-style-type: none"> <li>Fibres were non-treated or treated in NaOH solution</li> <li>Three fibre contents: 2, 5, 10%</li> <li>Alkali treatment modified the fiber structure: rough surfaces that provided good adhesion and interlocking with the polymer matrix</li> <li>Treatment to fibres improve the performance</li> </ul>	46





Filler	Modification/ Compatibilization	Processing Method	Functions/Properties	Ref.
			<ul style="list-style-type: none"> <li>Mechanical properties were improved overall and the improvement varies depending on the type of properties and fibre content</li> </ul>	
<b>Rice straw</b>	None	Extrusion	<ul style="list-style-type: none"> <li>The modulus of elasticity and modulus of rupture reached maximum values of 4917 MPa and 25.2 MPa at 1250 kg/m<sup>3</sup> of composite density with 60% and 50% fiber ratio, respectively</li> </ul>	47
<b>Agave fibres</b>	Grafting poly(methyl methacrylate) (PMMA) on fibre surfaces	Hand layup → hot pressing	<ul style="list-style-type: none"> <li>20% fiber content gives optimum mechanical properties</li> <li>Different fiber dimensions (particle, short and long fibers) on the mechanical properties of the composites has also been investigated</li> <li>Particle reinforcement gives better mechanical properties than short and long fiber reinforcement</li> <li>Grafting PMMA on fibres improved composite properties</li> </ul>	48
<b>Coconut shell</b>	Maleated PS	Compounding in an internal mixer	<ul style="list-style-type: none"> <li>Removing air in EPS by heating at 140 C, internal mixer</li> <li>Addition of MAPS improved the flexural properties by promoting the adhesion between cellulose and PS</li> </ul>	45



## 3 RECYCLING OF POLYSTYRENE

In the plastic industry, the most frequently used materials are identified by a number, *i.e.*, resin identification code (RIC), which is designed to help sorting of plastics during recycling. PS is number 6 in this system and the full list is in Table 6.

Table 6. Resin identification numbers and the corresponding polymers.

RIC	Polymer Name	Abbreviation
1	Polyethylene terephthalate	PET
2	High density polyethylene	HDPE
3	Polyvinylchloride	PVC
4	Low density polyethylene	LDPE
5	Polypropylene	PP
6	Polystyrene	PS
7	Others	Other

Plastic recycling can be categorized in four grades named from primary to quaternary. Their meanings are explained in Table 7. According to this, chemical recycling is a tertiary recycling process.

Table 7. Categories of plastic recycling.<sup>49</sup>

Name	Description	Examples
Primary Recycling	Recycling of plastics during the production process where the materials are easily identified and not contaminated	Close-loop processing scraps on production lines
Secondary Recycling	Recovery of used plastics. The materials are contaminated, and proper sorting is required. The properties of the recycled materials deteriorate	Extrusion of post-consumer waste
Tertiary Recycling	Degrading polymers to smaller molecules via chemical, physical, or biological processes	Chemical recycling of PS
Quaternary Recycling	Recovery of energy	Incineration of plastics

Both rigid PS and EPS can be recycled by physical method, *i.e.*, secondary recycling. During this process, the EPS needs to be degassed and some examples were given in section 2.1. PS can also be degraded to produce styrene monomers. In this section, the focus will be on the latter case. Even though the depolymerization of PS can be promoted via chemical or thermal approaches, it will be called chemical recycling PS in the following text.

### 3.1 Introduction of Polystyrene Chemical Recycling

A special feature of PS is that it can be chemically recycled. That is to say, PS can be decomposed through pyrolysis to give styrene monomer as the major product. Compared to that, other widely used addition polymers, *e.g.*, polyethylene (PE) and polypropylene (PP) generate diverse products during



pyrolysis and cannot be chemically recycled.<sup>50</sup> The recycled styrene monomer can be polymerized to produce PS products that are safe for food and medical application.<sup>9</sup> The circle of chemical recycling of PS is illustrated in Figure 7.

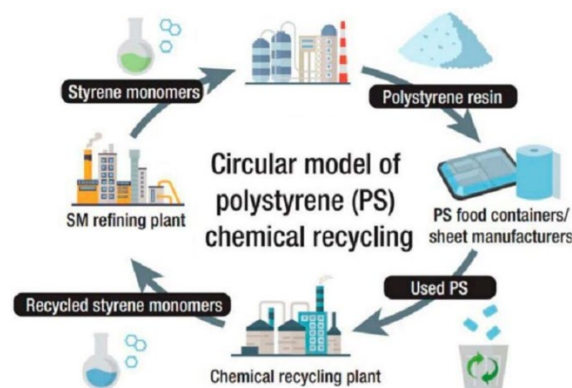


Figure 7. Diagram of chemical recycling model for PS.<sup>51</sup>

The pyrolysis of PS involves heating plastics at temperatures of 300-900 °C in the absence of oxygen, through which the long polymer chains decompose to low molecular weight oil, gaseous products, and char.<sup>52,53</sup> The pyrolysis of PS can be realized through different methods. A summary of these processes and their advantages are shown in Table 8.

Table 8. Various methods for PS pyrolysis.<sup>52</sup>

No.	Methods	Advantages
1.	Classical heating method	
	a. Batch reactor	high conversion efficiency
	b. Fixed bed reactor	simplicity in design
	c. Fluidized bed reactor	<ul style="list-style-type: none"><li>• Yield of liquid products is more than 90wt.%</li><li>• Formation of gas and coke is relatively insignificant</li></ul>
	d. Conical spouted bed reactor	<ul style="list-style-type: none"><li>• Allows better blending of charge</li><li>• Can treat large particles with density disparity</li></ul>
	e. Free-fall reactor under vacuum	Produces important liquid chemicals such as benzene, toluene, and naphthalene besides styrene monomer and valuable gaseous output
2.	Microwave-assisted pyrolysis	<ul style="list-style-type: none"><li>• Extremely rapid heating</li><li>• Higher production rate</li><li>• Low production cost</li><li>• Energy saving</li></ul>
3.	Catalytic pyrolysis	<ul style="list-style-type: none"><li>• Decreases the operating temperature</li><li>• Reduces the heat energy requirement</li><li>• Favors the industrial application of pyrolysis</li><li>• Produce final products of commercial significance</li></ul>
4.	Solvent-assisted pyrolysis	<ul style="list-style-type: none"><li>• High heat and mass transfer rates</li><li>• Reduces operating temperature</li><li>• Higher liquid yields</li></ul>



The pyrolysis of PS is a complicated process affected by many parameters, which need to be carefully controlled with the main objectives of high production yield and low energy cost. The important parameters include feedstock quality, temperature, reactor design, catalyst, and solvent.<sup>52,54</sup> An example setup for PS pyrolysis is shown in Figure 8. Catalysts are required to improve the efficiency of PS pyrolysis. Typical types of catalyst include bentonites,<sup>55</sup> acid and base,<sup>56</sup> zinc bulk,<sup>57</sup> and fluid catalytic cracking (FCC) catalyst<sup>58</sup>. Of the products from PS pyrolysis, the gas components are mainly ethylene and propylene, whereas the liquid phase is composed of various chemicals.<sup>59</sup> The chemicals in the liquid phase can be categorized into three groups according to their boiling points, and the main products are shown in Table 9.

Table 9. Main products from PS pyrolysis and their fractions based on boiling points.<sup>59</sup>

Fractions	Pyrolysis Products
Low boiling fraction (BP: < 200 °C)	Benzene
	Toluene
	Ethylbenzene
	Xylene
	Styrene (monomer)
	$\alpha$ -Methylstyrene
	Others
Medium boiling fraction (BP: 200-350 °C)	1,2-Diphenylethane
	1,3-Diphenylpropane
	2,4-Diphenyl-1-butene (Dimer)
	2,4-Diphenyl-1-pentene
	Others
High boiling fraction (BP: >350 °C)	2,4,6-Triphenyl-1-hexene (Trimer)
	Others

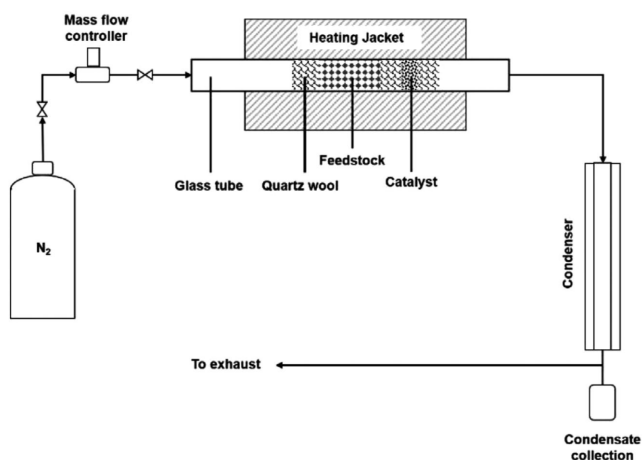


Figure 8. A typical setup for PS pyrolysis.<sup>54</sup>

In addition to producing styrene monomer as the main product, recent studies showed that other valuable products can be produced from PS by controlling the reaction conditions, a process called upcycling. It has been reported that PS can be degraded to formic acid, benzoic acid, and acetophenone



via a photo-acid-enabled protocol.<sup>60</sup> Another interesting study investigated degradation of PS to aromatics using UV light, followed by converting the intermediate to diphenylmethane, a much more valuable chemical than styrene and aromatics. This process has been tested at 1-kg scale in laboratory.<sup>61</sup>

## 3.2 Current market

Owing to plastic waste pollution concerns, recycling of plastics has been gaining more and more attention by both industry and government. Chemical recycling of PS has been commercialized over the last decade. There have been companies established around the world for the scale-up of chemical recycling PS, and the first commercial plant opened by Agilyx in 2018.<sup>62</sup> After that, more plants, pilot or commercial scales, were built around the world and a summary of the major ones are listed in Table 10.

Table 10. Current major commercial PS chemical recycling plants.

Region	Company	Location	Pilot/Commercial Scale	Ref.
<b>North America</b>	Agilyx	Tigard, Oregon, USA	10 US tons per day	62
	Pyrowave	Montreal, QC, Canada	200 tons per year	63,64
	Polystyvert	Montreal, QC, Canada	9,000 tons per year*	65
	GreenMantra	Brantford, ON, Canada	1,000 tons per year	66
	Ineos Styrolution /Agilyx	Channahon, Illinois, USA	100 tons per day	67
<b>East Asia</b>	Denka/Toyo	Chiba, Japan	3,000 tons per year*	51
<b>Europe</b>	Sulzer	Antwerp, Belgium	30,000 tons per year*	68,69
	Trinseo-Synova-Worley	Tessenderlo, Belgium	15,000 tons per year*	70,71

\*Under construction

The three companies located in Canada, Pyrowave, Polystyvert, and GreenMantra are using different technologies for depolymerization, which are microwave, dissolution using an essential oil, and thermo-catalytic depolymerization, respectively. GreenMantra's technology can also recycle PE and PP to produce wax as additives for polymer processing.

## 4 OUTLOOK

As reviewed in this report, plastic pollution may cause serious environmental impacts. PS is one of the most widely used plastics for applications such as packaging materials, containers, construction, and electronic housing. Owing to the chemistry of PS, it can be converted to its monomer with identical properties as the ones from fossil sources. Thus, PS can be fully circulated in the economy.

Regarding applications of wood derivatives in PS for composites, it has two aspects in perspective of chemical recycling PS.

1. Is there any difference between the composites made from the virgin PS and the chemically recycled PS?
2. Would the presence of wood derivatives in PS composites affect the chemical recycling process?



For the first question, the chemically recycled PS possesses identical properties as the virgin one. Therefore, the processing and properties of such composites are expected to be the same, regardless of the source of PS. For the second question, monomer purity is critical for PS production, and all companies doing chemical recycling PS claim that their technology can remove contaminants and additives, even small molecule substances, from waste PS. Thus, PS composites containing wood derivatives should be able to be fully recycled chemically.

The challenges for compounded wood derivatives with PS may come from the following points:

- PS is an inexpensive plastic. Addition of any materials into PS must be feasible in terms of economy and/or functionality.
- Recycling PS composite material should be viable in terms of TEA and LCA.
- One of the properties of rigid PS is transparency, which is normally desired for applications like containers. PS composites filled with wood derivatives will lose this feature. Therefore, acceptance of non-transparent or colored products might be a market issue.
- The typical processing temperature of PS is 215-260 °C, approaching the point where cellulose fibres start to decompose. The processing conditions of PS composites with wood fibres need to be carefully controlled.
- PS is not compatible with wood derivatives, thus proper compatibilization, e.g., pretreatment of the fillers or addition of coupling agents, is necessary for better performance.

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