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Lignocellulosic fibre mediated rubber composites: An overview

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ABSTRACT

The rising concern towards the reduction in the use of petroleum-based, non-renewable resources and the need for more versatile polymer-based composite materials have led to increasing interests on natural polymer composites filled with natural organic fillers, i.e. coming from renewable and biodegradable sources. This paper reviews wood flour and other lignocellulosic fibres filled rubber composites, including cellulosic rubber composites, cellulosic thermoplastic elastomers, nanocellulose based rubber nanocomposites, with the aims at providing the most state of the art information for directing further scientific research, possible commercialization and design of cellulosic rubber composites. It has been found that 1) the surface properties of natural cellulose, hence the compatibility and interface of the natural cellulose and matrix rubber/plastics, are crucial for the successful development of the composites, such, physical and chemical modification and additives have been widely attempted to improve the incompatibility and poor interfacial adhesion between the filler and matrix; 2) the curing characteristics, mechanical properties, thermal stability and morphologies of the composites are complex but closely related to not only the interfacial properties, but also the compositions (e.g. the concentration of cellulosic materials) and other processing parameters; 3) the nature of hydrophilic cellulosic and hydrophobic matrix rubber and/or plastics requires an accurate introduction of coupling agent, one end of its structure shall be compatible to hydrophilic and the other to hydrophobic. The reviews on the main paths and results of study on the advanced nanocellulose reinforced rubber nanocomposites and sandwiches indicate much potentials and needs for further in-depth studies.

1. Introduction

Lignocellulosic fibres and rubber can both be from natural resources. Due to their excellent flexibility, elasticity, electrical property, resistance to crack propagation, water and most fluid chemicals, building tackiness, and other superior mechanical and chemical properties, both natural and synthetic rubbers in vulcanized form are used to produce various rubber products, such as hoses, cushions, gloves, seals, tyres, belts, diving gear, chemical and medicinal tubing, electrical instruments [1–4]. In the traditional vulcanization process of rubbers, carbon black (CB) and silica are commonly employed as fillers for the benefits of improving strength, stiffness and weather resistance, which lead to a special class of material known as composites [4–7]. In spite of the capabilities of CB and silica as the reinforcements in rubber composites, both CB and silica are non-degradable and consume amount of energy for their production. CB is also a petrochemical based product. Therefore, rubber manufacturers are searching for new reinforcing fillers which are renewable, biodegradable, inexpensive, light-weight, and readily available.

Over the past decade, the use of wood and other cellulosic fibres as fillers or reinforcements to replace CB and silica in the production of rubber composites has attracted much attention. The specific nature of these natural products, namely abundance, low density and cost, renewable character, high specific strength and modulus, harmlessness and biodegradability, has facilitated their association with natural and synthetic rubbers to elaborate composite materials [8–11], and these bio-based degradable composites are becoming new generation materials [12]. Currently, wood and other lignocellulosic fibres mediated rubber composite materials can be categorized into wood flour and other cellulosic fibres filled composites, nanocellulose reinforced natural rubber nanocomposites and wood flour-rubber laminates.

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Wood flour and other lignocellulosic fibres can be easily incorporated into rubber compound with other ingredients to fabricate bio-based rubber composites through the standard rubber processing operations, such as compress moulding, injection moulding and extrusion [4]. Although these composites provide good strength and dimensional stability during fabrication, the main issues, namely the compatibility between rubber and wood or fibre, and their interfacial adhesion, should be and are yet to be addressed, since the properties of the lignocellulosic rubber composites are dominated by the interaction between the filler and matrix [13–15], especially the surface property of the filler is polar and hydrophilic, while the matrix, rubber is generally non-polar and relatively hydrophobic [16]. Numerous surface modifications aiming at improving the miscibility and compatibility between rubber and wood or natural fibre have been carried out recently [17–19]. Generally, the modification strategies fall into two categories, namely physical treatment and chemical treatment.

Thermoplastic elastomer is another class of hybrid composites, which has better functional performance than rubber since the properties can be tailored by changing the ratio of the rubber to the plastic in the composite [20–22]. There have been several investigations that combine wood flour or other cellulosic fibres with thermoplastic elastomer to produce novel rubber composites [23–25]. The incorporation of biomass not only enhances some specific properties of the novel composite, such as tensile strength and elongation at break, but also reduces the consumption of the petroleum product (plastics) and improves the biodegradability of the resultant materials [26]. However, the degradability of cellulosic composite against environmental attack is an indispensable issue needed to be investigated when considering its outdoor applications. Although the degradability of thermal elastomers (i.e. EPDM), synthetic fibre (i.e. melamine and aramid fibres) filled thermal elastomers and cellulosic plastic composites has been extensively studied [27–32], the degradability of cellulosic rubber composites and cellulosic thermal elastomers has not been virtually conducted.

Cellulose is the most abundant organic polymer on earth. The cellulose content of cotton fibre is about 90% and that of wood and other green plants is about 40–50%. The cellulose for industrial use is usually obtained from wood pulp and cotton, although cellulose is recently also produced from other resources. Nanofibrillated cellulose has been used in nanocomposites in combination with various matrices due to its typically ultra-high strength and environmental friendliness [33,34]. During the past five years, nanocellulose was explored as a reinforcing phase in natural rubber latex for rubber nanocomposites fabrication [4]. The nanocellulose provides the rubber nanocomposite with superior mechanical strength as it is expected, meanwhile, it increases the rate of degradation of rubber in soil when disposed at the end of life [35].

In the past few years, in order to deal with the increasing amount of waste tyre rubber, some investigations on novel wood-rubber composites have been carried out, although the results showed that an addition of used tyre rubber resulted in a reduction of internal bond of rubber-wood particleboard [36–38].

It is evident that the interest in biobased composites continues to increase due to the environmental benefits, especially in pure biobased products, such as lignocellulosic rubber composites, both constituents of which can be sourced from renewable materials. For both industry and research communities, it has therefore become of importance to overview and assess the state of the art for both commercial exploitation and further development, on which unfortunately little review is available. The main goal of this paper is to study the formulation, properties and performance of various wood and other cellulosic fibres mediated rubber composites, hence provides fundamental basis for further research in the area and enables the optimization of the composites for possible industrializations.

2. Lignocellulosic fibre rubber composites

2.1. Formulation of cellulosic rubber composites

2.1.1. Modification of lignocellulosic fibres and rubber matrix

Numerous reports are available on wood flour and other lignocellulosic rubber composite (Table 1). It is well known that the main disadvantage of the lignocellulosic fibre reinforced rubber composite is the poor compatibility between the hydrophilic wood flour or other natural fibres and hydrophobic rubber matrix, which leads to the unsatisfactory mechanical properties of the composite. The modification of the surface characteristics of the wood flour and other cellulosic fibres is needed in particular in order to formulate a reasonable composite [39,40]. Various approaches, including physical and chemical treatments, have been attempted for modifying the surface of cellulosic fibres and improving the compatibility or interface properties. They can be grouped and further discussed as follows:

Radiation treatment is one of the physical techniques used in wood- or natural fibre-plastic composite, enabling significant physical and chemical changes as well as changes in surface structure and surface energy of the wood or fibres. The common radiation treatments are the UV radiation, electron beam radiation, gamma radiation and corona treatment [41–43]. A typical treatment of gamma radiation is known to deposit energy on the wood or fibre in the composite and radicals were then produced on the cellulose chain by hydrogen and hydroxyl abstraction, ruptures of some carbon–carbon bonds and chain scission. Simultaneously, peroxide radicals are generated when matrix polymers are irradiated in the presence of oxygen. These active sites in both fibre and matrix produced by the gamma radiation result in the better

Table 1
Wood flour and other cellulosic fibres filled rubber composites.

Filler	Rubber matrix	References
Wood flour/powder	Natural rubber Ethylene-propylene diene rubber Styrene butadiene rubber Waste tyre rubber	[63–70]
Oil palm flour	Natural rubber Epoxidized natural rubber	[71–75]
Olive husk powder	Carboxylated nitrile butadiene rubber	[59]
Hemp hurd powder	Styrene butadiene rubber	[58]
Linoleum flour	Acrylonitrile butadiene rubber	[76]
Peanut shell powder	Natural rubber	[77,78]
Coconut shell powder	Natural rubber	[12]
Pistachio shell powder	Natural rubber Styrene butadiene rubber	[79]
Rice husk powder	Natural rubber Styrene butadiene rubber	[24,50,80]
Rattan powder	Natural rubber	[81,82]
Oil palm fibre	Natural rubber	[2,83,84]
Bamboo fibre	Natural rubber	[85]
Coir fibre	Natural rubber	[51,86]
Hemp fibre	Natural rubber Ground tyre rubber	[87,88]
Bagasse fibre	Natural rubber	[89]
Sisal fibre	Natural rubber Tyre rubber	[51,84,90]
Jute fibre	Natural rubber	[44]
Cotton fibre	Natural rubber	[60]
Newsprint paper	Natural rubber Nitrile butadiene rubber	[91]
Microcrystalline cellulose	Styrene butadiene rubber Poly-butadiene rubber	[92]