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Thermogravimetric behavior of natural fibers reinforced polymer composites—An overview

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ABSTRACT

Natural fibers obtained from plants, known as lignocellulosic fibers are environmentally friendly alternatives for synthetic fiber, as polymer composite reinforcement. Applications of natural fiber composites are expanding in many engineering areas, from civil construction to automobile manufacturing. In recent years, a considerable number of scientific and technological works, including review papers, were dedicated to the characterization and properties of natural fibers and their composites. The mechanical behavior and the fracture characteristics are usually the most investigated and reviewed themes for the purpose of comparison to corresponding polymer composites reinforced with synthetic fibers, mainly fiberglass. The thermal behavior is also of practical interest for conditions associated with temperatures above the ambient, as in fire damage, curing or process involving heating procedures. In fact, several works also assessed distinct thermal responses, particularly in terms of thermogravimetric properties of natural fiber polymer composites. As no general review was conducted so far on the thermogravimetric (TG) behavior of these materials, this article presents an overview limited to temperature effects related to the loss of mass by means of TG analysis and the related derivative, DTG, for different polymer composites reinforced with the most common and relevant lignocellulosic fibers.

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1. Introduction

In the past few decades, environmental issues concerning global scale pollution and climate changes renewed the interest in natural materials including cellulose-rich fibers extracted from cultivated plants, also known as lignocellulosic fibers, and their polymer composites [1–11]. Actually, natural fibers are considered environmentally friendly not only for their saving in process energy, which is an unavoidable problem for synthetic fibers, but also for their renewable and biodegradable characteristics. Both natural fibers and their polymer composites, if incorporated with a reasonable amount of fiber, are neutral with respect to CO_2 emissions that cause the earth greenhouse effect, a major responsible for global warming [12]. In fact, at the end of these “green” materials life cycle, the release of CO_2 due to combustion or atmospheric degradation will be balanced by the content assimilated during the fiber’s plant biological growth [1].

In addition to environmental, economical and social benefits, some physical and mechanical properties found in natural fibers and related composites [1–11] represent important technical advantages over synthetic fibers. This is of special relevance when compared to glass fiber polymer composites (fiberglass) today extensively used in a wide variety of engineering systems in most industrial sectors. Fiberglass is heavier, difficult to machine and cannot be recycled even by incineration in thermoelectric plants. It also has potential health hazards posed by fiber particulates [13,14]. An expansion of industrial applications of natural fiber composites is currently taking place aiming to substitute the traditional uses of fiberglass in building construction, packaging, sports, electrical parts, medical prosthesis and automobile components [10]. In particular, the automobile industry is increasingly adopting natural composites in numerous interior and exterior components [15–19]. Among the technical advantages of natural fibers and their composites, it stands the relatively low density, which results in improved specific properties of significant interest to the automobile industry. For instance, the energy consumption of 9.6 MJ/kg to produce a flax fiber mat, including cultivation, harvesting and fiber separation, is significantly lower than the energy of 54.7 MJ/kg to produce a glass fiber mat [17]. Moreover, for composite processing, a non-abrasive natural fiber is associated with less damage to tools and molding equipments as well as a relatively better finishing in comparison to glass fiber. Additionally, owing to an inherent flexibility and easy delamination with respect to polymer matrices, natural fiber composites are also tougher and resist impact loads without shattering [9].

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Comparatively, important drawbacks are exhibited by natural fibers that, to some extent, also affect their polymer composites [1–11]. As biomaterials, natural fibers have dimensions that are limited by anatomical restrictions with accentuated statistical dispersion. Consequently, all lignocellulosic fibers present a technically undesirable feature associated with heterogeneous characteristics in contrast to uniform synthetic fibers fabricated within precise dimensional values. Another shortcoming is the fact that natural fibers are hydrophilic and tend to develop a weak bonding with hydrophobic polymers normally used as composite matrices [1–4,9–11]. Surface modifications [20,21] may improve the fiber adherence to a polymer matrix and thus increase both the composite mechanical and thermal resistance [1–4]. A surface treatment, however, results in additional cost and decreases the economical competitiveness of the natural fiber composites. The thermal stability of any natural fiber composite may also impose limitations in applications at temperatures that cause degradation of the fiber organic structure. In principle, the temperature not only degrades the structure but also affects most properties of the natural fiber composites. A complete understanding of these effects requires a review on the composite basic thermogravimetric, i.e., weight loss with increasing temperature characterization. In spite of many specific works, no overview on thermal behavior has been presented so far and, therefore, this is a motivation for this overview article.

2. Thermal degradation of lignocellulosic fibers

Among the review articles and books [1–11] on properties and structural characteristics of natural lignocellulosic fibers and their polymer composites, only that one of Nabi Sahed and Jog [2] presented a section related to general aspects of the thermal stability. Works referred in that section [22–29] served for the brief summary on the state of art discussed by these authors [2]. In two conference articles on thermal analysis of lignocellulosic materials, Nguyen et al. [22,23] reviewed the effect of temperature on cellulose, hemicellulose, lignin, and other carbohydrates as well as different types of wood. It was indicated that the thermal decomposition of cellulose begins to occur at 210–260 °C by dehydration followed by major endothermic reaction of depolymerization with DTG peaks that vary from 310 to around 450 °C. Hemicellulose was found to decompose at a maximum of 290 °C and up to 150 kJ/mol for activation energy while lignin would thermally decompose with peaks from 280 to 520 °C and up to 229 kJ/mol for activation energy.

The cellulose, hemicellulose and lignin degradation caused by temperature was considered by Nabi Sahed and Jog [2] a crucial aspect for the thermal stability of natural fiber reinforced polymer composites. In the case of a thermoset matrix, this is a limiting factor for choosing the curing temperature, while in thermoplastic matrix composites, a limitation for extrusion temperature. Thermal protection of the fiber was attempted by grafting of acrylonitrile on jute [25], which increased the degradation temperature to 280 °C, and on sisal [26], by lowering both the rate of degradation as well as the total weight loss. The maximum rate of degradation was also reduced in wood flour by previous treatment with phosphonate [25]. Works on the effect of thermal degradation of wood flour incorporated polymer composites [27,28] showed that mechanical properties deteriorate with increasing temperature. Toughness and bending strength were the most affected. This was attributed to changes in the surface chemistry of the wood flour, which might affect the bonding with the polymeric matrix and impair the composite properties. In another work on wood flour incorporated polypropylene composites, similar loss in properties were reported after extrusion at 250 °C [29]. As a general comment, Nabi Sahed and Jog [2] mentioned that thermal degradation of natural fibers also results in production of volatiles at processing temperatures above 200 °C. This can lead to porous polymer composites with inferior mechanical properties. They stated that the real challenge for the scientists is to improve the thermal stability of these fibers, so that they can be used with engineering polymers for better performance and thus widen the applications of natural fiber composites.

As final remarks, the reader should find relevant to know that the thermogravimetric behavior of natural fibers apparently bears a correlation with their chemical constituents. In fact, the TG/DTG curves of common lignocellulosic fibers such as jute, sisal, wood and cotton display similar aspects that could be correlated to the thermal decomposition of their main constituents. Three stages of weight loss are associated with the TG curve. A first stage, up to about 200 °C, corresponds to a maximum weight loss of 10% and is followed by a second stage with more than 70 wt% of loss, up to about 500 °C. The final third stage extends to the usual ending test temperature at about 800 °C in association with a loss that may reach 20 wt%. The corresponding DTG curves display maximum rate of thermal decomposition peaks as well as shoulder and tail peaks that could be ascribed to the fiber's constituents. Table 1 exemplifies important TG/DTG parameters obtained from figures of quoted review papers [7,22]. The values shown in this table are similar to others reported for distinct natural fibers but may vary depending on treatments performed on the fiber [7]. According to Nguyen et al. [22], the weight loss and DTG peak in first stage can be attributed to water loss. The thermal degradation of the main lignocellulosic constituents of the fiber begin to occur at the onset of the second stage. The main DTG peak in the second stage can be ascribed to the cellulose decomposition, while the shoulder peak to the hemicellulose and the tail peak to the end of lignin decomposition. The weight remaining in the third stage could be assigned to char or other products from decomposition reactions.

Another point worth discussing is the influence of different TGA atmospheres on the thermogravimetric results of natural fibers. In principle, two distinct atmospheres, inert (helium and nitrogen) and oxidative (air and oxygen) may be used. Moreover, as gases conduct heat at different rates, thermograms obtained in nitrogen may be significantly different from those obtained in helium. Under inert atmosphere, the thermal degradation of cellulose results in a main DTG peak associated with the formation of macromolecules containing rings bearing double bonds [22]. In an oxidative atmosphere there is partial overlapping of this peak with the exothermic peak corresponding to the oxygen reaction with the cellulose. As a consequence, the main DTG peak is shifted to lower temperatures in oxidative atmosphere as compared to the inert one. For instance, the maximum rate of

Table 1
Thermogravimetric parameters of common natural fibers [7,22].

Natural fiber	1st stage weight loss (%)	1st stage DTG peak (°C)	2nd stage onset T_0 (°C)	2nd stage weight loss (%)	2nd stage DTG shoulder (°C)	2nd stage DTG main peak (°C)	2nd stage DTG tail (°C)	3rd stage weight loss (%)
Jute [7]	8	60	260	89	290	340	470	3
Sisal [7]	9	52	250	76	275	345	465	15
Wood [22]	2	107	290	85	270	367	400	13
Cotton [7]	4	55	265	91	280	330	410	5

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