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Effects of nanofiber orientations on the fracture toughness of cellulose nanopaper

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ABSTRACT

Cellulose nanopaper exhibits superior mechanical properties with both high strength and toughness, and the crack bridging mechanism of nanofibers makes the most significant contribution to its fracture toughness. In this paper, we investigate the fracture toughness of a mode-I crack in cellulose nanopaper by using a modified crack-bridging model. Different from previous crack-bridging models, we account for the effect of nanofibers inclined to the crack surfaces. Particular attention is given to the dependence of fracture toughness on the orientation distribution of nanofibers in the cellulose nanopaper. We use a cohesive law to account for the interfacial shear stress of nanofibers, which involve self-healing of hydrogen bonds at their interfaces. Two representative orientation distributions are considered, in which nanofibers are aligned or randomly oriented, respectively. The theoretical results agree well with relevant experiments. This work helps understand the structure–property relationship of cellulose nanopaper and design other fiber-reinforced nanocomposites.

1. Introduction

Cellulose nanofibers, derived from natural materials such as wood and bamboo, have attracted much attention due to their promising applications in bio-based nanomaterials [1–5]. Cellulose nanopaper consisting of a porous network of cellulose nanofibers suggests a bottom-up material design strategy to combine high strength and toughness [6–11]. As yet, however, the mechanical potentials of cellulose nanopaper have not been fully attained because of a lack of understanding of its property–microstructure relation.

Considerable efforts have been dedicated recently toward exploring how to improve the mechanical properties of cellulose nanopaper. For example, nanopaper with a preferred orientation of cellulose nanofibers was fabricated via cold drawing in the wet state [12–14]. The results showed that an increased degree of nanofiber alignment enhances the mechanical stiffness and strength of cellulose nanopaper along the alignment direction. This enhancement effect was also observed in macrofibers made of cellulose nanofibers aligned by wet-stretching [15–18]. Håkansson et al. [19] prepared cellulose filaments by utilizing a surface-charge-controlled gel transition combined with hydrodynamically induced fiber alignment. They showed that those filaments with less aligned fibers exhibit lower stiffness and tensile strength but higher strain at break compared with those made of more aligned fibers. More recently, an anisotropic cellulose nanopaper with highly aligned cellulose nanofibers by delignification and mechanical pressing of natural wood was fabricated by Zhu et al. [20]. They found that the alignment of cellulose nanofibers can dramatically increase the strength and toughness of the resulting anisotropic nanopaper compared with its counterpart without alignment. These

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Nomenclature

δ	crack opening displacement	f	probability density function of nanofiber orientations
θ	orientation angle of a nanofiber	F_b	bridging force
λ	crack-bridging zone length	K_I^0	intrinsic fracture toughness
λ_s	maximal crack-bridging zone length	K_I^∞	stress intensity factor in the far field
σ_∞	tensile stress in the far field	K_I^b	stress intensity factor induced by bridging nanofibers
σ_b	bridging stress	K_I^c	fracture toughness of nanopaper
τ_s	interfacial shear strength between nanofibers	l	average nanofiber length
A	fiber orientation distribution parameter	R	nanofiber radius
E	elastic modulus of nanopaper	V_f	volume fraction of nanofibers

experiments evidenced that nanofiber alignment contributes significantly to the mechanical properties of cellulose nanopaper, and when all nanofibers are aligned along the tensile direction, the full mechanical potentials of cellulose nanopaper can be achieved. To date, however, there has been a lack of theoretical understanding of the nanofiber alignment effects on the mechanical properties of cellulose nanopaper. Fundamental research in this regard would help to design cellulose nanopaper with desirable mechanical performance.

In this paper, therefore, the effect of nanofiber orientations on the fracture toughness of cellulose nanopaper is elucidated by using a modified crack-bridging model. The relation between the fracture toughness and microstructure is established for cellulose nanopaper with either fully aligned or randomly oriented nanofibers.

2. Model and method

2.1. Crack-bridging model

Cellulose nanopaper usually has a porous network microstructure consisting of randomly distributed nanofibers [7–11]. Its mechanical properties can be tuned over a wide range by adjusting the sizes and orientations of nanofibers. In particular, cellulose nanopaper comprising fully aligned nanofibers has anisotropic properties, with the highest strength and toughness along the nanofiber direction [12–14,20]. To examine the effect of nanofiber orientations, therefore, we here compare the fracture toughness of cellulose nanopaper in the following three typical cases of uniaxial tension: (i) all nanofibers are aligned along the tensile direction, (ii) all nanofibers are aligned along a direction deviated from the tensile stress with an angle θ, and (iii) the nanofibers are randomly oriented, as shown in Fig. 1. Other cases of nanofiber orientations can be analyzed analogously.

During loading, the nanofibers embedded in the cellulose nanopaper may slide relatively to each other. As the load increases beyond a threshold, a crack may initiate and propagate. During crack propagation, nanofibers behind the crack tip can be partly

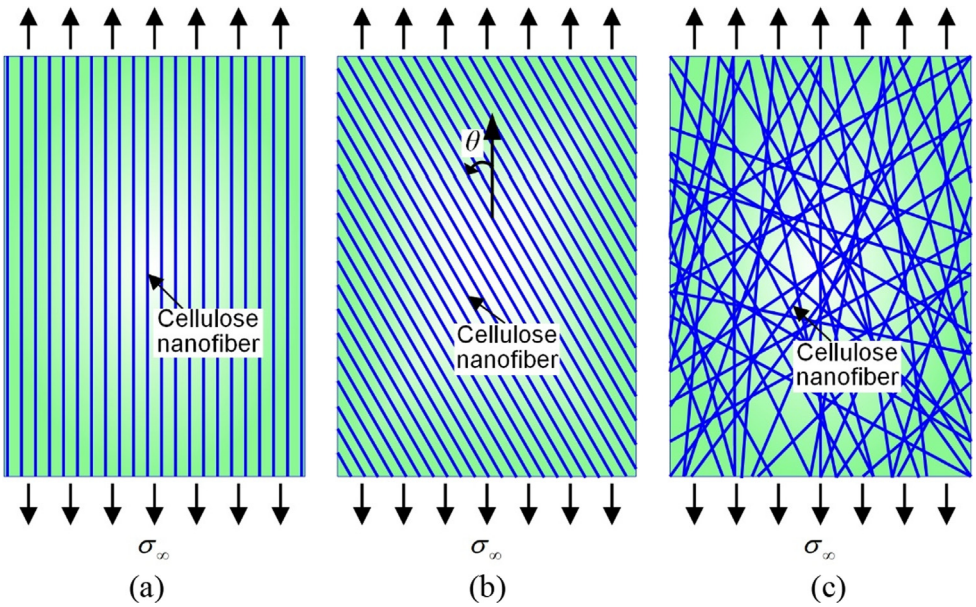

Fig. 1. Schematic of (a) a nanopaper under with fully aligned nanofibers along the tensile stress σ_∞, (b) a nanopaper with fully aligned nanofibers and subjected to tension with an angle θ measured from the fiber direction, and (c) a nanopaper with random nanofibers.