

# Surface behavior and film formation analysis of sisal fiber coated by poly(methyl methacrylate) ultrathin film

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## Abstract

Admicellar polymerization was used to modify a sisal fiber surface with poly(methyl methacrylate) (PMMA) in order to improve the compatibility between the sisal fiber and the surrounding polymeric matrix in a composite. The effect of the amount of monomer (methyl methacrylate) and initiator (sodium persulfate) on the hydrophobicity behavior and PMMA film formation of the admicellar-treated sisal surface was studied. The increase in the hydrophobicity of the admicellar-treated sisal fiber was examined by flotation testing, moisture absorption, and electrostatic charge or zeta ( $\zeta$ ) potential. The amount of PMMA film formed on the sisal surface was investigated by the weight loss of the admicellar-treated sisal extracted by acetone and chloroform; and the thermal degradation was studied by thermogravimetric analyses. The admicellar-treated sisal could float on the surface of water for longer than half an hour, and its moisture absorption decreased. The  $\zeta$  potential of its surface also showed a significant change compared to the untreated sisal. The results from the weight loss indicated that the amount of PMMA formed on the sisal fiber surface depended on the amount of monomer and initiator. The Fourier transform infrared spectrum of the admicellar-treated sisal showed the characteristic peaks of PMMA and the scanning electron micrograph of the treated sisal was clearly different from the untreated sisal, confirming that there was a thin film coating on the admicellar-treated sisal fiber.

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## 1. Introduction

Sisal fiber accounts for almost half of the total production of commercially used natural fibers. In addition, sisal fibers, which are extracted from the leaves of the sisal plant (*Agave sisalana* Perr), are widely grown in the tropical zone, including Thailand. At present, sisal fiber is mainly used as rope, twine, cords, upholstery, padding and mat making, fishing nets, fancy articles, etc. During the past decade, the use of sisal fiber as an economical and environmentally friendly reinforcement for polymeric composites has raised great interest. However, sisal, which is a cellulose fiber, typically has poor interfacial adhesion with a hydrophobic polymer matrix and low moisture resistance, leading to a decrease in the durability of the composites. Recent

developments in chemical and thermal methods for the modification of the sisal fiber surface in order to enhance the adhesion between the sisal fiber and the surrounding polymeric matrix and to reduce water absorption were reviewed by Li et al. [1]. The chemical composition and mechanical properties of sisal fiber are summarized in Table 1 [2]. Currently, there are several new methods for improving the natural fiber–polymer compatibility; for example, alkaline treatment [3,4], silane treatment [4,5], and graft copolymerization of monomer directly on the surface [5–7]. Surface treatment by admicellar polymerization is a new surface modification method proposed as a competitive method for compatibility enhancement in the composite. The admicellar polymerization process is generally carried out in an aqueous surfactant solution system in order to form the admicelle on the substrate surface which acts as a two-dimensional container for polymer film formation from an organic monomer.

In general, cellulose fibers contain a large number of hydroxyl groups in their chemical structure, as shown in Fig. 1a, and in an aqueous system, the polar groups with a negative charge

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Table 1  
Chemical composition and mechanical properties of sisal fiber [2]

Chemical composition	
Cellulose (%)	67–78
Lignin (%)	8.0–11.0
Hemicellulose (%)	10.0–14.2
Moisture content (%)	11.0
Mechanical properties	
Tensile strength (MPa)	468–640
Young's modulus (GPa)	9.4–22.0
Elongation at break (%)	3–7

occur through interactions with water molecules, as illustrated in Fig. 1b [8]. The charge of the sisal surface can be investigated by measuring its  $\zeta$  potential in an aqueous solution at different pH values. In order to control the surface charge and/or the hydrophilic/hydrophobic character of the surface, surfactants which are adsorbed at the solid–liquid interface can be used. The adsorption of a cationic surfactant causes a decrease in the negative  $\zeta$  potential down to zero, which is the iso-electric point. In order to form the admicelle of the surfactant on the cellulose fiber, a cationic surfactant can be used. However, pH and counter ions can increase the amount of surfactant adsorbed on the fiber surface to form the admicelle needed for the admicellar polymerization process.

Admicellar polymerization is an innovative method, first investigated by Wu et al. [9]. It can be used to improve the compatibility at the interface between different materials. In previous work, admicellar polymerization has been used to improve the adhesion in polymeric composites, such as precipitated silica with polystyrene [10] and the copolymers of styrene–butadiene and styrene–isoprene [11], and glass fibers with polystyrene [12] and the copolymer of isoprene–styrene [13]. There are also many researchers applying the admicellar polymerization technique to other areas for various purposes by coating different polymers on different materials [9,14–19]. For reinforcement modification, all studied reinforcements were inorganic materials, while many types of natural cellulose fiber have received much interest for use as the reinforcement with the benefit of being environmentally friendly. In this work, admicellar polymerization was used in order to modify the natural cellulose fiber for possible future use in a natural fiber–polymer composite.

Admicellar polymerization generally consists of three main steps: admicelle formation, monomer adsolubilization, and polymer formation, as illustrated in Fig. 2. The method makes use of the formation of a surfactant bilayer or admicelle on a substrate at a surfactant concentration just below the critical micelle concentration (CMC). In the outer surfactant layer, the amphiphilic molecules are oriented with the ionic head groups in contact with the aqueous solution, while the long-hydrophobic

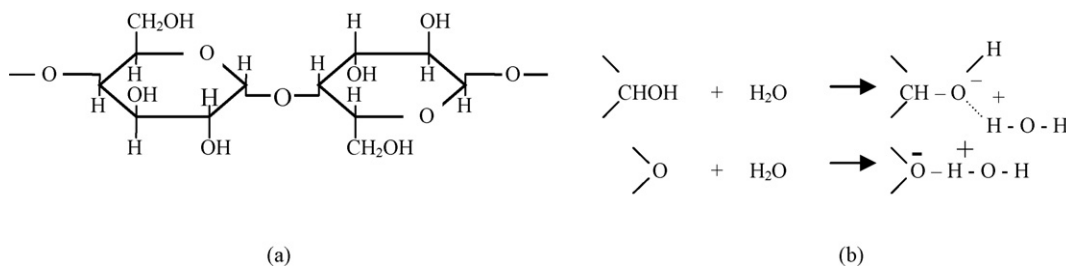


Fig. 1. Cellulose structure: (a) chemical structure and (b) interactions with water molecules.

### 1. Admicellar Formation



### 2. Monomer Adsolubilization



### 3. Polymer Formation



Fig. 2. The admicellar polymerization process.

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