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## **ACCEPTED MANUSCRIPT**

# Underwater superoleophobicity cellulose nanofibril aerogel through regioselective sulfonation for oil/water separation

Feifei Sun<sup>1,2</sup>, Wei Liu<sup>1</sup>, Zhaoxia Dong<sup>2</sup>, Yulin Deng<sup>1,\*</sup>

<sup>1</sup>School of chemical & Biomolecular Engineering, Renewable Bioproducts Institute, Georgia Institute of Technology, Atlanta, GA, 30332, USA

<sup>2</sup>Institute of Enhance Oil Recovery, China University of Petroleum, Beijing, China.

#### **Abstract**

Cellulose nanofiber (CNF) aerogel regioselectively modified by sodium periodate oxidation and consecutive sodium sulfite sulfonation has been fabricated. The aerogel exhibits underwater superoleophobicity ( $\theta_{oil} > 150^{\circ}$ ) characteristic for various oils, which could be used as a filter for oil/water separation. Compared with originals, the charge density of sulfonated CNF increased from -39.8 mmol/kg to -325 mmol/kg, which prevented the aggregation in water suspension and resulted in better dispersion morphology of nanofibers in the final aerogel after freezing-drying. Therefore, modified aerogel with a good hierarchical structure and high charge density was able to trap more water molecules onto its voids once pre-wetted, which is energetically unfavorable for the oil to replace the water molecules. As a result, the sulfonated aerogel displayed superoleophobicity characteristic underwater thus nonpolar oil phase was completely repelled while the water easily penetrated the as-prepared aerogel. The sulfonated CNF aerogel maintained high oil-water separation efficiency by simply filtration. The oil content in filtrated water was always less than 300 ppm (0.03%), and kept a stable recyclability even after 20 cycles, which could be practically used as a filter for oil/water.

#### 1. INTRODUCTION

Expanding industrial oily wastewater and frequent oil spill accidents have caused increasing

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