# Viscoelastic interfaces comprising cellulose nanocrystals and lauroyl ethyl arginate for enhanced foam stability

Agnieszka Czakaj<sup>a</sup>, Aadithya Kannan<sup>b</sup>, Agnieszka Wiśniewska<sup>c</sup>, Gabriela Grześ<sup>d</sup>, Marcel Krzan<sup>a</sup>,

Piotr Warszyński<sup>a</sup>, Gerald G. Fuller<sup>b</sup>

corresponding author. Email address: ncczakaj@cyf-kr.edu.pl

a Jerzy Haber Institute of Catalysis and Surface Chemistry PAS, Kraków, Poland

b Department of Chemical Engineering, Stanford University, Stanford, USA

c Institute of Physical Chemistry, Polish Academy of Sciences, Warsaw, Poland

d Department of Chemistry, Jagiellonian University, Kraków, Poland

# Supporting information

## **Bulk viscosity**

Shear viscosity measurements were performed by Malvern Kinexus Pro rotational rheometer with cone - plate geometry with diameter 50 mm, angle 1 degree, and 0.3 mm of a gap. The experiments were conducted in controlled shear stress mode with constant temperature 298K. All concentrations were prepared twice, sonicated for a time of 10 min and measured. The range of shear stress was 0.01 to 1.25 Pa. Assuming the proportional relation for shear stress and shear rate for Newtonian fluid, the dynamic viscosity was calculated by extrapolation to zero shear rate. The results are collected in Table S1.

Table S1. Bulk viscosity of the investigated mixtures of CNC nanoparticles (0.3% wt.) with LAE

	Zero shear rate viscosity	
CNC 0.3%	0.00136 Pa s	
LAE 0.005% - CNC 0.3%	0.00142 Pa s	
LAE 0.01% - CNC 0.3%	0.00135 Pa s	
LAE 0.015% - CNC 0.3%	0.00165 Pa s	

## CNC particle size and zeta potential

Size of CNC nanoparticles was measured by dynamic light scattering with Malvern Nano ZS instrument. Each measurement was repeated three times and standard error from 3 measurements is given.

Zeta potential of all LAE – CNC mixtures was measured by laser Doppler velocimetry with Malvern Nano ZS instrument. Each measurement was repeated three times. No viscosity correction was applied. The average error (standard deviation) was 5 mV maximum.

The results of measurements are given in Table S2.

Table S2. The changes of the hydrodynamic diameter and zeta potential of CNC nanoparticles on the addition of LAE.

	Mean diffusion coefficient [µm <sup>2</sup> /s]	Hydrodynamic diameter [nm] (polydispersity index)	Zeta potential [mV]
LAE	-	-	70
CNC 0.6% wt.	5.08	96 (0.63)	-44
LAE 0.004% - CNC 0.3% wt.	5.61	88 (0.56)	-44
LAE 0.006% - CNC 0.3% wt	6.36	77 (0.52)	-50
LAE 0.008% - CNC 0.3% wt.	5.59	88 (0.50)	-
LAE 0.015% - CNC 0.3% wt.	2.65	187 (0.82)	-45

## Lauroyl ethyl arginate



Fig. S1 Lauroyl ethyl arginate molecular structure (pubchem.ncbi.nlm.nih.gov/compound/188214)

#### Atomic force microscopy imaging

Atomic Force Microscopy (AFM) images were obtained using a Dimension Icon AFM (Bruker, Santa Barbara, CA) working in QNM<sup>®</sup> mode with SCANASYST-AIR tips (nominal spring constant of 0.4 Nm<sup>-1</sup>).

Cellulose nanocrystals were deposited layer-by-layer on a silicon wafer treated with piranha solution and with aqueous polyethyleneimine solution 0.1 g/100 mL (MW 750 000) to form positive anchor layer for negatively charged LAE-CNC species.

Figures S2 and S3 illustrate AFM images of dry film of cellulose nanocrystals deposited from 0.6% wt. Solution and dry film of cellulose nanocrystals deposited from LAE- CNC (0.015% : 0.3% wt.) mixture.







Fig. S3. Dry film of cellulose nanocrystals deposited from LAE - CNC (0.015% : 0.3% wt.) mixture layer-by-layer at polyethyleneimine – coated Si wafer.

## Scanning electron microscopy imaging

A drop of cellulose nanocrystals suspension (initial concentration 1 % wt.) was dried directly on the microscopic stage in atmospheric conditions. Samples were imaged without additional modification with Scanning Electron Microscopy (SEM) LEO 1450VP, Electron Microscopy Ltd., Cambridge, U.K.



Fig. S4. The images of 3D structure formed with CNC nanoparticles after drying the drop of suspension.

# Foaming



Fig. S5. Foams produced by double syringe method from LAE – CNC 0.3% mixtures. Left: 0.004% wt. LAE, Middle: 0.006% wt. LAE, Right 0.015% wt. LAE. Different starting point of foaming should be taken into account.

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Fig. S6. Fluid film of the bubble with cellulose nanocrystals assembled at the air/liquid interface (zoom).