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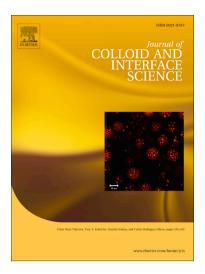
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### **ACCEPTED MANUSCRIPT**

# Cationic Polyacrylamide induced nanoparticles assembly in a cellulose nanofiber network

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#### **Abstract**

Polyacrylamides of different molecular weight, charges and dosages allow to control the retention and distribution of nanoparticles (NPs) in composites, and optimise composite properties and functionality. Our aim is to evaluate the effect of high molecular weight (13MDa) cationic polyacrylamide (CPAM) charge and dosage on SiO<sub>2</sub> (74 nm) NP's assembly in cellulose nanofibers composites.

Engineered cellulose/SiO<sub>2</sub> composites were investigated by SEM, SAXS and DLS. SEM images show the local area retention of NPs into the cellulose matrix. SAXS provides an average NPs distribution and inter-NPs distance over complete volume of composite. DLS gives the hydrodynamic radius of CPAM adsorbed onto SiO<sub>2</sub> NPs in a suspension.

SAXS analysis reveals a structure conformation made of spherical SiO<sub>2</sub> NPs core of diameter 74 nm surrounded by a CPAM polyelectrolyte shell 2.5 nm thick. Surprisingly, CPAM induced assembly of SiO<sub>2</sub> NPs with constant inter-nanoparticle distance, which is irrelevant of polymer charge density. However, NPs retention in the cellulose fiber network increases with CPAM dosage. The assembly mechanism is governed by the balance of electrostatic and steric forces following CPAM coverage onto NPs and the inter-nanoparticle CPAM bridging conformation. This maintains the constant inter-nanoparticle distance and the assembly of NPs in the cellulose network.

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