Nano-fibrillated cellulose aerogels: How density affects its properties

Deeptanshu Sivaraman[a,b,*], Shanyu Zhao[a], Gilberto de Freitas Siqueira[c], Gustav Nyström[c], Marco Lattuada[b], Matthias M. Koebel[a], Wim J. Malfait[a]

[a] *Empa – Building energy materials and components, Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland*

[b] *Department of Chemistry, University of Fribourg, Fribourg, Switzerland*

[c] *Empa – Wood and Nano cellulose laboratory, Swiss Federal Laboratories for Materials Science and Technology, Dübendorf, Switzerland*

*deeptanshu.sivaraman@empa.ch

ABSTRACT

Aerogels have exceptional properties that open-up various application areas. Their largest field of application is in buildings as a thermal insulation material, owing to its ultra-low thermal conductivity (λ). For well-studied examples such as silica aerogel, λ is minimal around 13 $mW.m^{-1}.K^{-1}$ at a density of ~0.110 g/cm^3 [1]. Biopolymers are potentially a more sustainable alternative to silica because of their renewable precursors. Cellulose is inexpensive, abundant and biodegradable. Kobayashi et al. [2] reported nanofibrillated cellulose (NFC) aerogels with a similar λ than silica aerogel, but at an order of magnitude lower density (18 $mW.m^{-1}.K^{-1}$ at 0.017 g/cm^3). In contrast, Plappert et al. [3] observed a minimum in λ at 0.087 g/cm^3, more in line to that for silica aerogel. Thus, the optimum density for cellulose aerogels to minimize λ remains controversial.

Here, we produce hydrogen-bonded NFC hydrogels using a novel and scalable procedure, by gas-phase acid gelation of TEMPO-oxidized NFC suspensions in water, solvent exchange to ethanol and supercritical CO_2 drying. The aerogels are transparent with BET surface areas of ~450 m^2/g and mesopore volumes of ~2 cm^3/g. Their λ decreases monotonically with increasing NFC concentration and density, from 37 to 28 $mW.m^{-1}.K^{-1}$ for densities from 0.005 g/cm^3 to 0.027 g/cm^3. Higher concentration-induced densities were not feasible due to limitations of the suspension viscosity. Higher densities were achieved by mechanical compression of lower density aerogels, resulting in a more pronounced reduction in λ with a minimum at 18 $mW.m^{-1}.K^{-1}$ at 0.065 $g.cm^{-3}$ and a traditional U-shaped curve. For a given density, λ is lower for samples prepared by mechanical compression, presumably due to the advantageous fiber alignment and pore anisotropy observed by SAXS analysis. Overall, our data are in line with those of Plappert et al. [3], albeit with lower λ values and better mechanical properties for a given density, but inconsistent with those of Kobayashi et al. [2].

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