

## Supporting Information

### **Surface Modification of Cured Inorganic Foams with Cationic Cellulose**

### **Nanocrystals and Their Use as Reactive Filter Media for Anionic Dye Removal**

Tuula Selkälä,<sup>a</sup> Terhi Suopajarvi,<sup>a</sup> Juho Antti Sirviö,<sup>a</sup> Tero Luukkonen,<sup>a</sup> Paivo Kinnunen,<sup>a</sup> Ana Luiza Coelho Braga de Carvalho,<sup>b</sup> and Henrikki Liimatainen<sup>a,\*</sup>

<sup>a</sup>*Fiber and Particle Engineering Research Unit, University of Oulu, P. O. Box 4300, FI-90014 Oulu, Finland*

<sup>b</sup>*Clausthal Technical University, Department of Mineral and Waste Processing, Walther-Nernst-Straße 9, 38678 Clausthal-Zellerfeld, Germany*

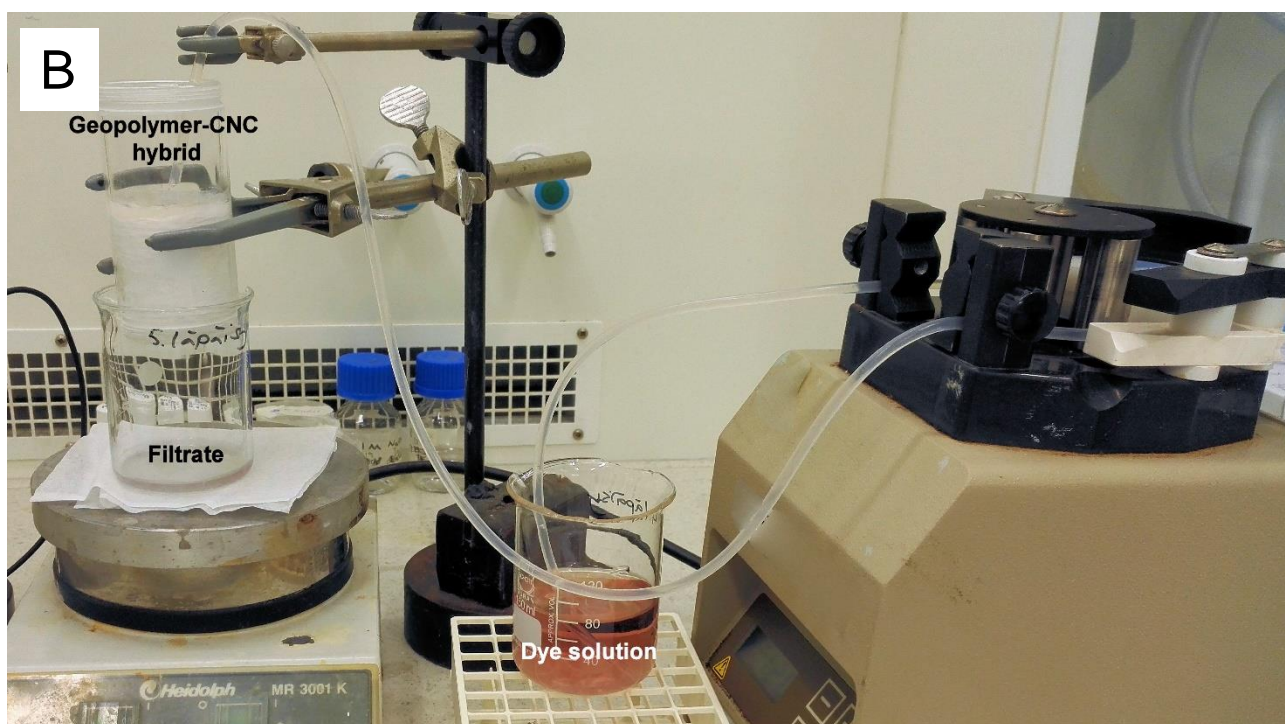
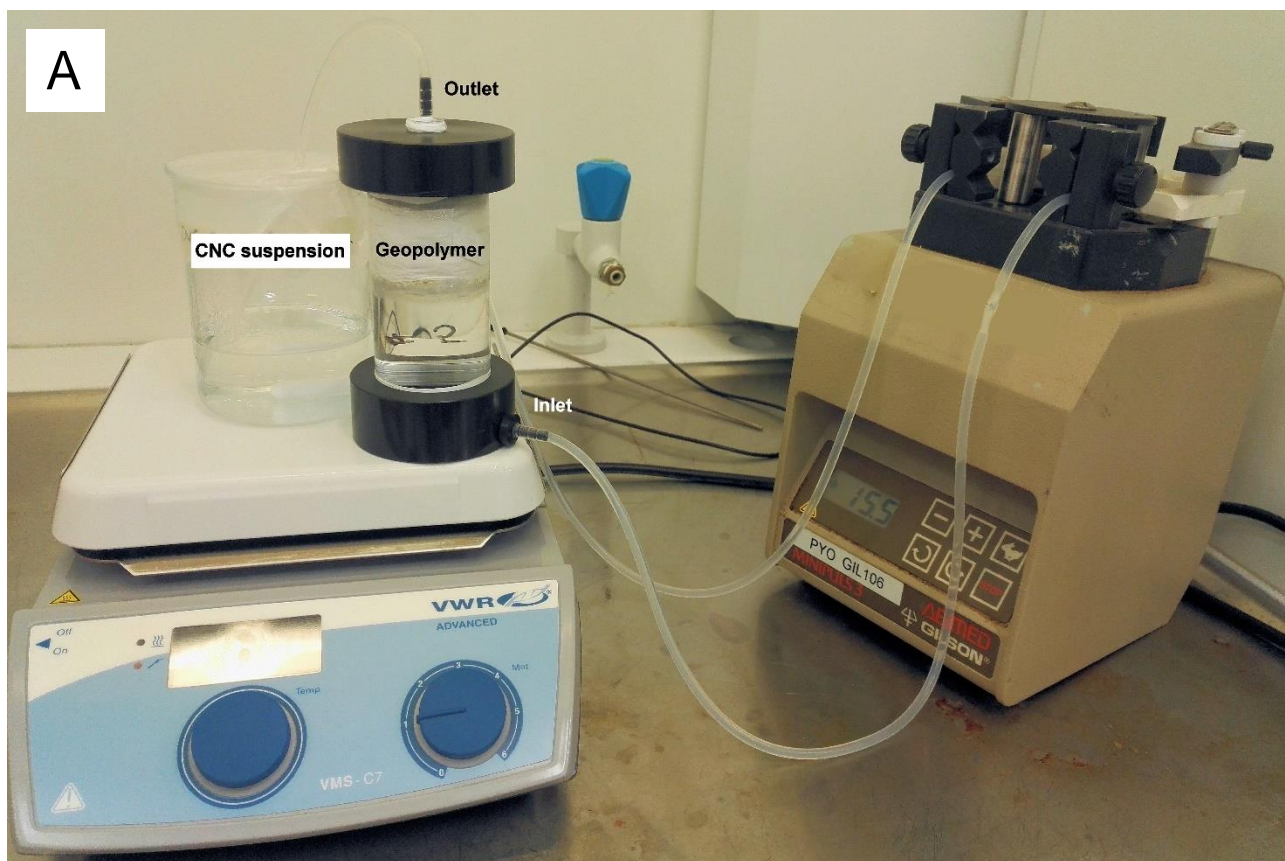
*\*Corresponding author; Tel: +358505659711. Email: henrikki.liimatainen@oulu.fi*

Number of pages: 9

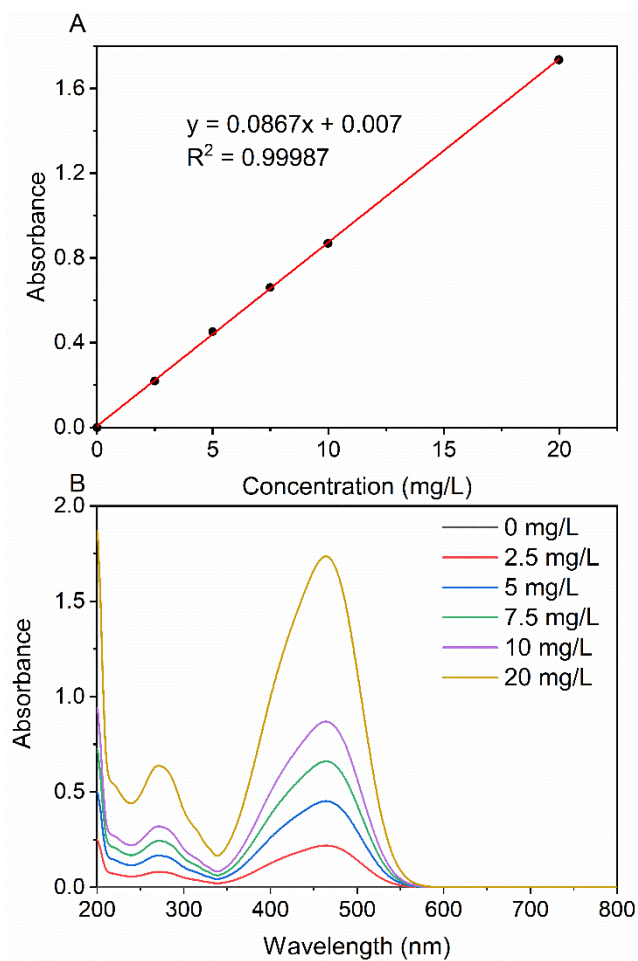
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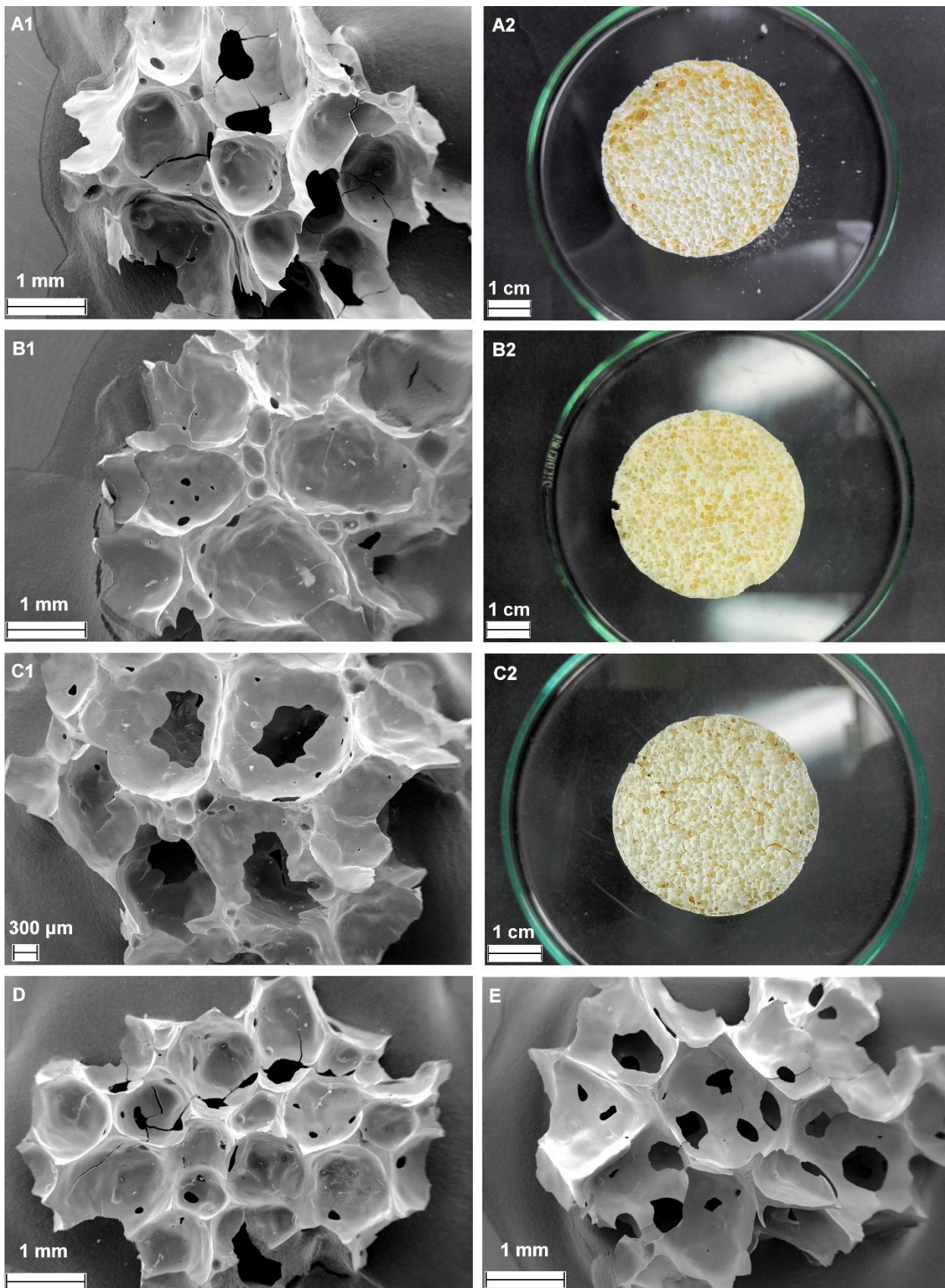


**Figure S1.** Set-ups used in the (A) CNC filtration and (B) dye adsorption experiments. The geopolymer samples in the photographs were shorter than what were used in the actual experiments reported in the article.

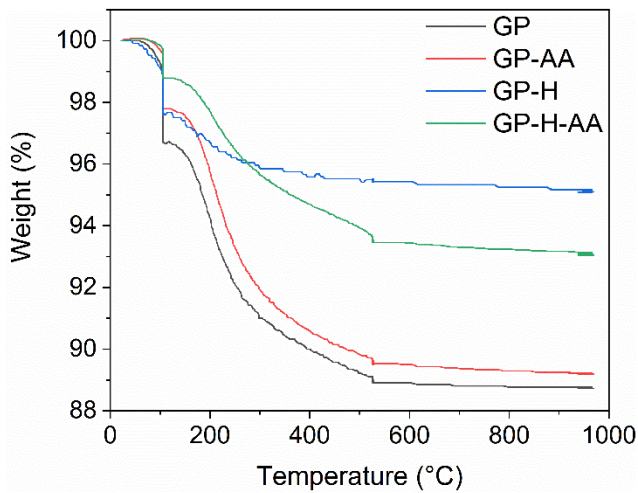


**Figure S2.** (A) Methyl orange calibration curve showing UV-vis absorbance as a function of solution concentration ( $\lambda_{\text{max}}$  464 nm, 0–20 mg/L in deionized water, pH 7). (B) UV-vis spectra of methyl orange in deionized water (0–20 mg/L, pH 7).

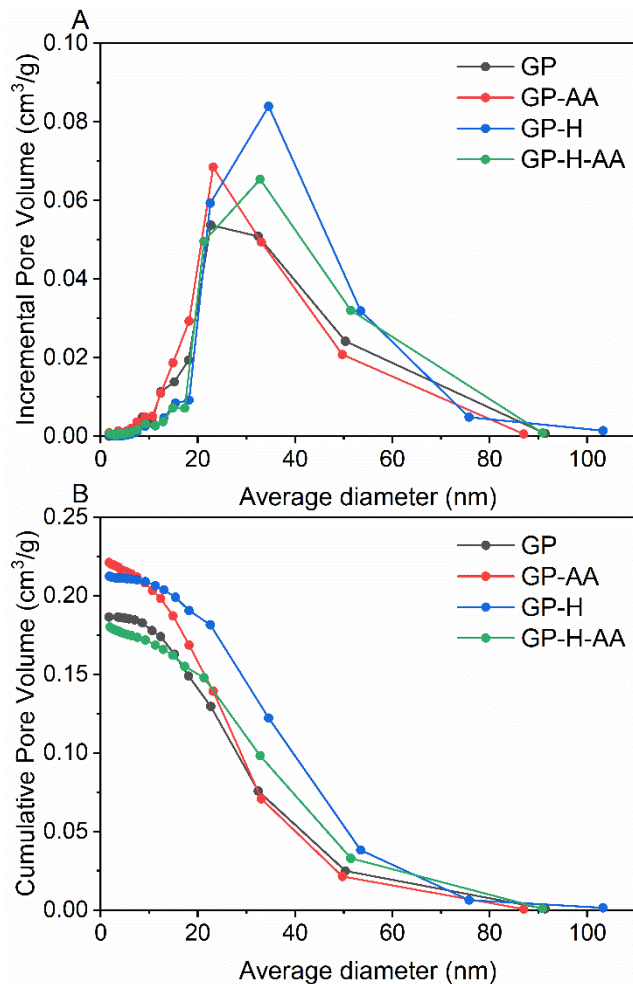




**Figure S3.** Field emission scanning electron microscope (FESEM) images and photographs of: (A1-A2) geopolymer (GP); (B1-B2) hydrothermally treated geopolymer (GP-HT); and (C1-C2) heat-treated geopolymer (GP-H) samples. FESEM images of (D) acid-washed (GP-AA) and (E) heat-treated acid-washed (GP-H-AA) geopolymers.



**Figure S4.** TGA curves of geopolymer (GP), acid-washed geopolymer (GP-AA), heat-treated geopolymer (GP-H) and heat-treated acid-washed geopolymer (GP-H-AA) samples under air atmosphere between 22–950 °C.



**Figure S5.** (A) Incremental and (B) cumulative pore volumes of geopolymer (GP), acid-washed geopolymer (GP-AA), heat-treated geopolymer (GP-H) and heat-treated acid-washed geopolymer (GP-H-AA) samples calculated from the BJH desorption data.

**Table S1.** Comparison of synthesis of geopolymer foams using a direct foaming method.

	Luukkonen et al. 2020	This manuscript 2020	
<b>Raw materials</b>			
Metakaolin (53.0 wt% SiO <sub>2</sub> , 44.5 wt% Al <sub>2</sub> O <sub>3</sub> )	x	x	
Sodium silicate solution (molar SiO <sub>2</sub> /Na <sub>2</sub> O ≈ 3.5, water content ≈ 64 wt%)	x	x	
NaOH pellets	x	x	
30% H <sub>2</sub> O <sub>2</sub>	x	x	
CTAB	x	x	
<b>Amounts</b>			
Alkali activator SiO <sub>2</sub> /Na <sub>2</sub> O molar ratio	1.2	1.6	
Metakaolin/alkali activator weight ratio	1.00 : 1.36	1.00 : 1.79	
Water content of fresh paste [wt%]	33	40	
H <sub>2</sub> O <sub>2</sub> content of fresh paste [wt%]	0.6	1.0	
CTAB content of fresh paste [mol/g]	1.947×10 <sup>-6</sup>	1.584×10 <sup>-6</sup>	
Curing	60 °C 4 h	60 °C 4 h	
<b>Geopolymer</b>			
SiO <sub>2</sub> /Al <sub>2</sub> O <sub>3</sub> molar ratio	3.24	GP	GP-H
Na <sub>2</sub> O/SiO <sub>2</sub> molar ratio	0.31	3.65	3.65
Na <sub>2</sub> O/Al <sub>2</sub> O <sub>3</sub> molar ratio	1.00	0.27	0.27
H <sub>2</sub> O/Na <sub>2</sub> O molar ratio	1.00	1.00	1.00
H <sub>2</sub> O/Na <sub>2</sub> O molar ratio	9.80	14.1	14.1
Total porosity (gas pycnometer) [%]	88.8	93.3	93.0

**Reference:**

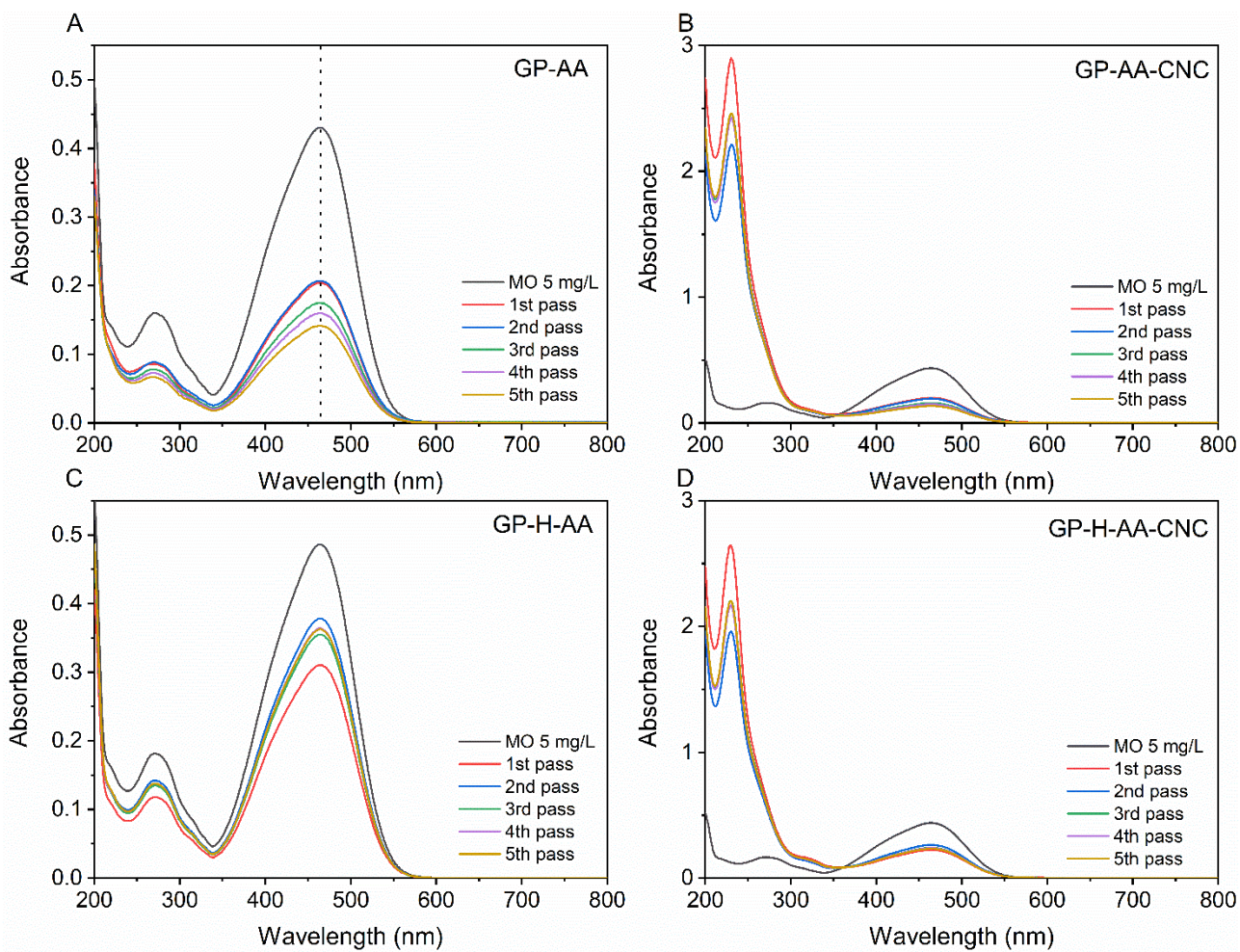
Luukkonen, T., Yliniemi, J., Sreenivasan, H., Ohenoja, K., Finnilä, M., Franchin, G., & Colombo, P. (2020). Ag-or Cu-modified geopolymer filters for water treatment manufactured by 3D printing, direct foaming, or granulation. *Scientific Reports*, 10(1), 1-14. <https://doi.org/10.1038/s41598-020-64228-5>



**Table S2.** Parameters used in the surface coverage calculations.

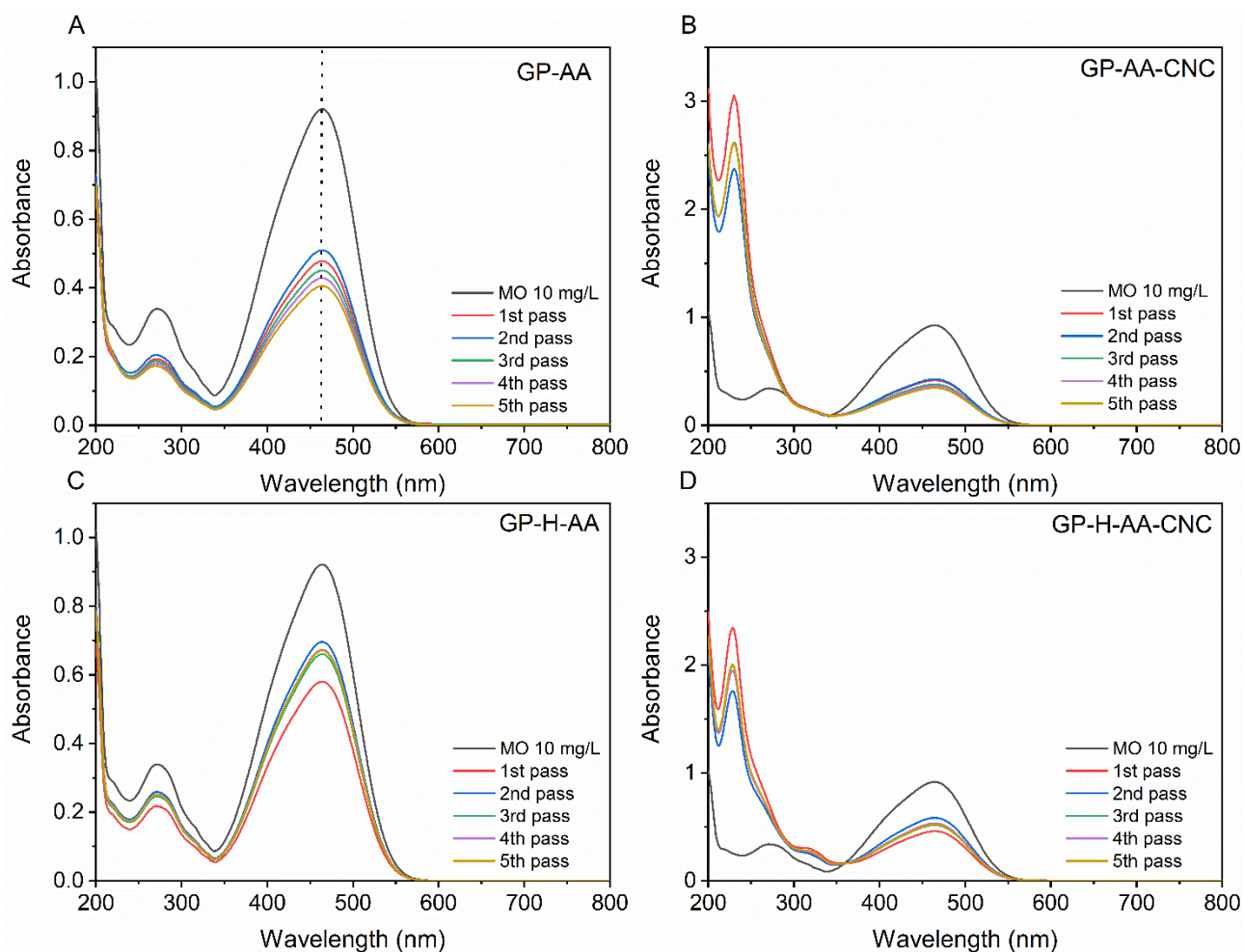
Parameter		Source	Value				Unit
Cellulose density	$\rho$	Assumption	1500				[kg/m <sup>3</sup> ]
CNC width	d	EFTEM images	$8.00 \times 10^{-9}$				[m]
CNC length	l	EFTEM images	$9.80 \times 10^{-8}$				[m]
Surface area of one longitudinal side of one CNC	$A_{\text{CNC}}$	Assumption: Rectangular shape, $A = d \times l$	$7.84 \times 10^{-16}$				[m <sup>2</sup> ]
Volume of one CNC	V	$V = d \times d \times l$	$6.27 \times 10^{-24}$				[m <sup>3</sup> ]
Mass of one CNC	$m_1$	$m_1 = \rho \times V$	$9.41 \times 10^{-18}$				[g]
			<b>GP-AA<sup>a</sup></b>	<b>GP-AA</b>	<b>GP-H-AA</b>	<b>GP-H-AA</b>	
Mass of CNC suspension	$m_2$	Experiments	600	600	600	600	[g]
Dry matter content of CNC suspension		Experiments	0.1	0.1	0.1	0.1	[% w/w]
Mass of CNCs in the suspension	$m_3$	$m_3 = 0.001 \times m_2$	0.6	0.6	0.6	0.6	[g]
Mass of CNCs adsorbed in the geopolymer	$m_4$	Experiments	0.297	0.274	0.243	0.346	[g]
Number of CNCs adsorbed in the geopolymer		$m_4/m_1$	$3.2 \times 10^{16}$	$2.9 \times 10^{16}$	$2.6 \times 10^{16}$	$3.7 \times 10^{16}$	pcs
Total CNC surface area in contact with the geopolymer	$A_{\text{tot(CNC)}}$	$A_{\text{tot(CNC)}} = (m_4/m_1) \times A_{\text{CNC}}$	24.8	22.8	20.3	28.8	[m <sup>2</sup> ]
Mass of geopolymer	$m_5$	Experiments	24.0	21.9	21.2	19.0	[g]
CNCs adsorption on geopolymers		$(m_4 \times 1000)/m_5$	12	13	11	18	[mg/g]
Geopolymer specific surface area	SSA	BET analysis	47.8	47.8	36.5	36.5	[m <sup>2</sup> /g]
Total surface area of geopolymer	$A_{\text{tot(geo)}}$	$A_{\text{tot(geo)}} = \text{SSA} \times m_5$	1145	1046	773	693	[m <sup>2</sup> ]
Surface coverage		$(A_{\text{tot(CNC)}}/A_{\text{tot(geo)}}) \times 100$	2.2	2.2	2.6	4.2	[%]
Average surface coverage			<b>2.2</b>		<b>3.4</b>		[%]

<sup>a</sup>There were two pieces of each geopolymer type. One was tested with 5 mg/L MO solution and the other with 10 mg/L MO solution.



**Figure S6.** UV-vis spectra of methyl orange solution (initial concentration 5 mg/L, pH 7) after filtrations through: (A) acid-washed geopolymer (GP-AA); (B) acid-washed geopolymer combined with CNCs (GP-AA-CNC); (C) heat-treated and acid-washed geopolymer (GP-H-AA); and (D) heat-treated and acid-washed geopolymer combined with CNCs (GP-H-AA-CNC).





**Figure S7.** UV-vis spectra of methyl orange solution (initial concentration 10 mg/L, pH 7) after filtrations through: (A) acid-washed geopolymer (GP-AA); (B) acid-washed geopolymer combined with CNCs (GP-AA-CNC); (C) heat-treated and acid-washed geopolymer (GP-H-AA); and (D) heat-treated and acid-washed geopolymer combined with CNCs (GP-H-AA-CNC).