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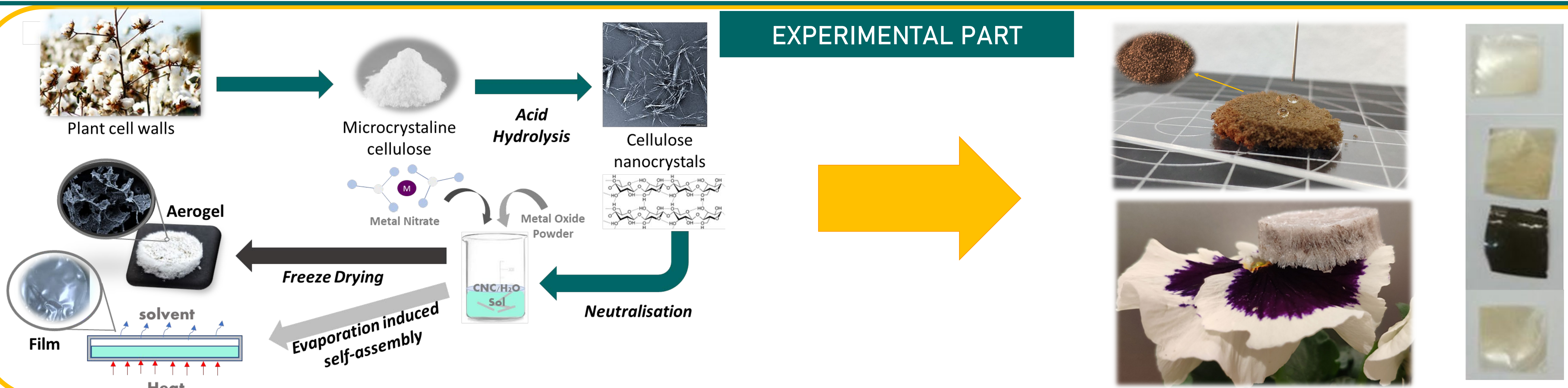
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INTRODUCTION

Production of **biodegradable materials** based on natural resources is a **growing area of interest** due to the serious problem of waste accumulation and plastic contamination in oceans and seas. Besides, naturally available materials in their nanometric scale like **nanocellulose** have attracted **great interest** during the last years, owing to their **low density, high specific surface, high aspect ratio, biocompatibility, abundance and wide functionalization possibilities** to obtain high value products with low impact. Among this functionalization possibilities, **transition metal containing nanocellulose** is an **interesting alternative for different application fields**.

This work reports on the **development of cellulose nanocrystals (CNC) based nanomaterials** (aerogels and films) **loaded with different metal oxide nanoparticle (MNP)** through scalable and **surfactantless methods**. Furthermore, CNC/MNP aerogels have been **reinforced by crosslinking** with other biopolymer, **sodium alginate (ALG)**. Morphological aspects and different **properties like wettability, disintegrability, UV-Visible transmittance and thermal stability and mechanical resistance** have been studied and compared.

EXPERIMENTAL PART



Morphology

AEROGELS

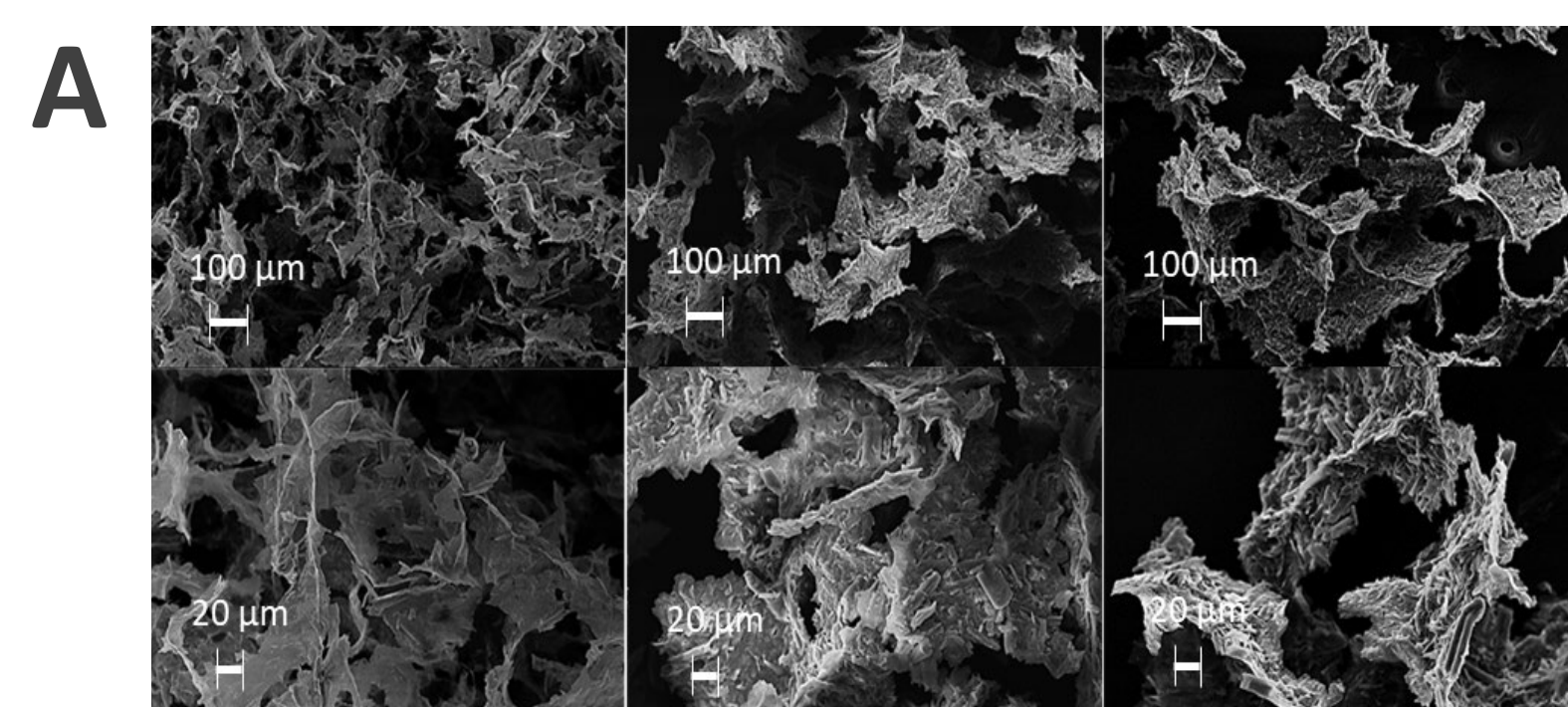
Wettability

RESULTS

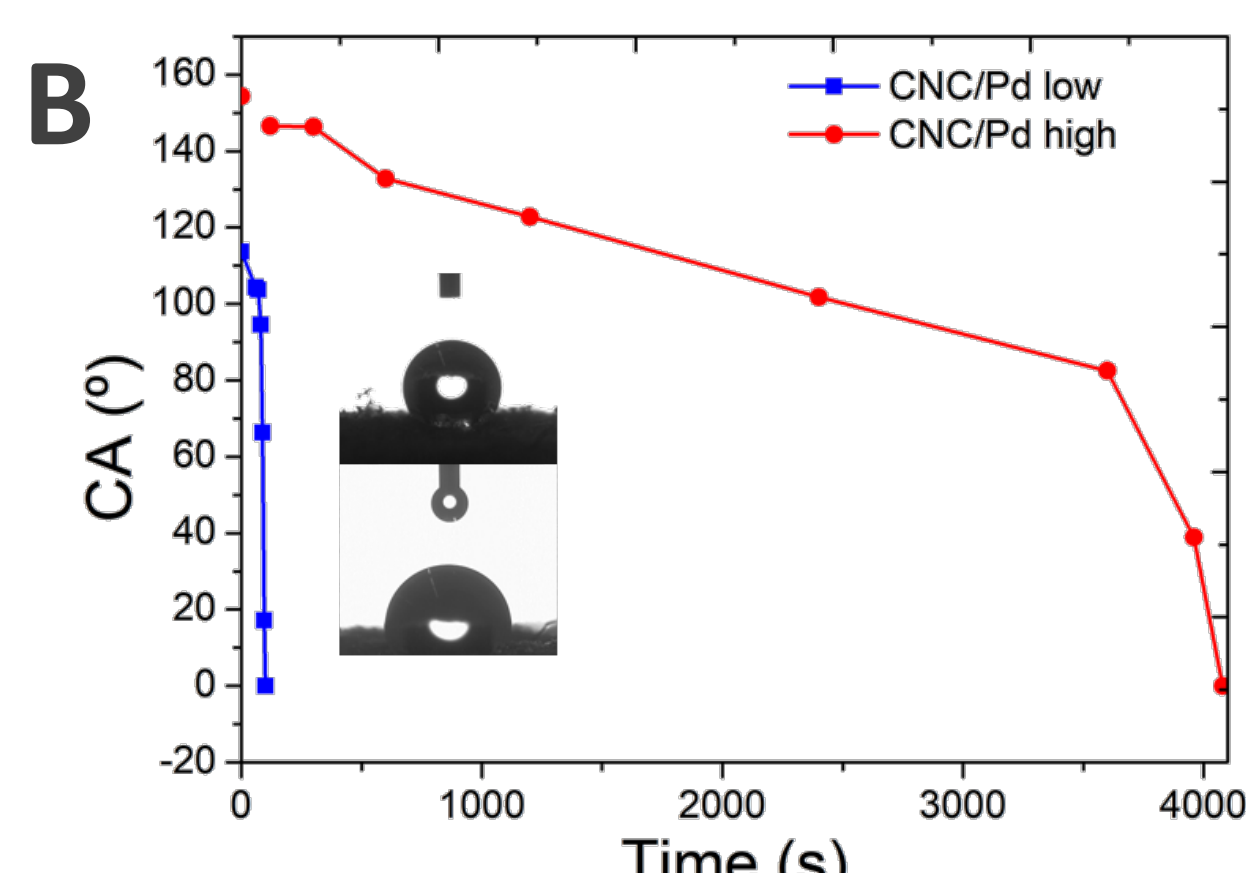
Morphology

FILMS

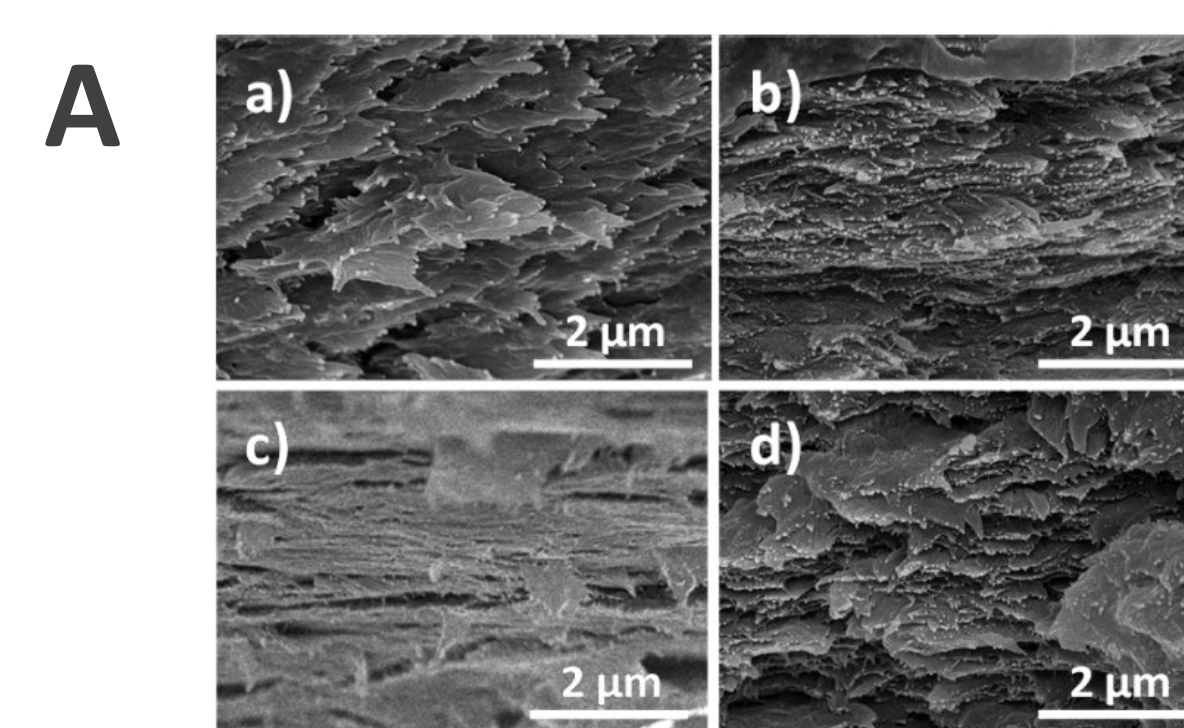
Disintegrability



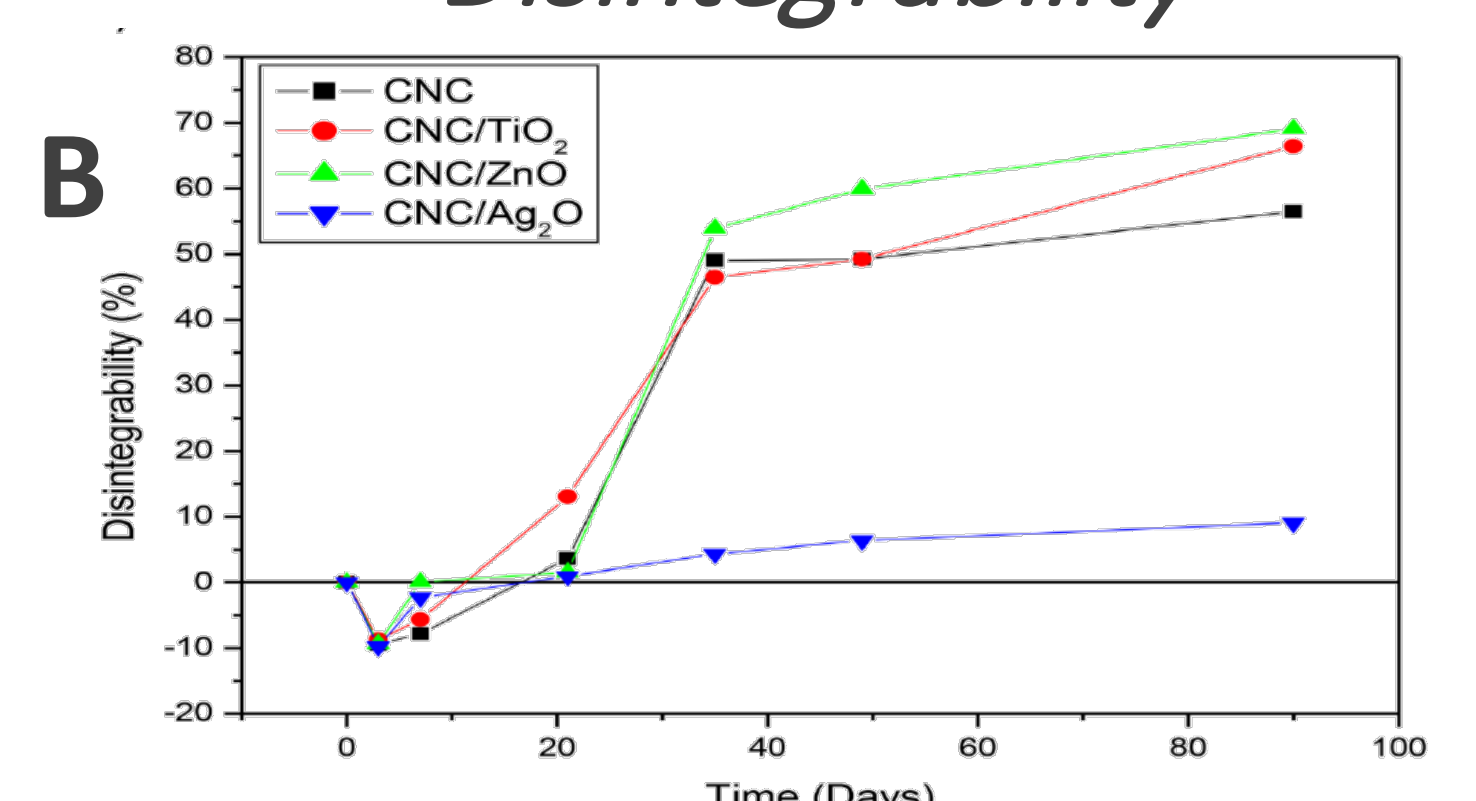
Obtained CNC, CNC/Cu and CNC/Pd aerogels show low density (12-20 mg/cm<sup>3</sup>) and high porous (98%) mesoporous structures.



Palladium loaded aerogel shows water contact angles increment with a hydrophobic behavior CA < 140° for CNC/Pd high samples

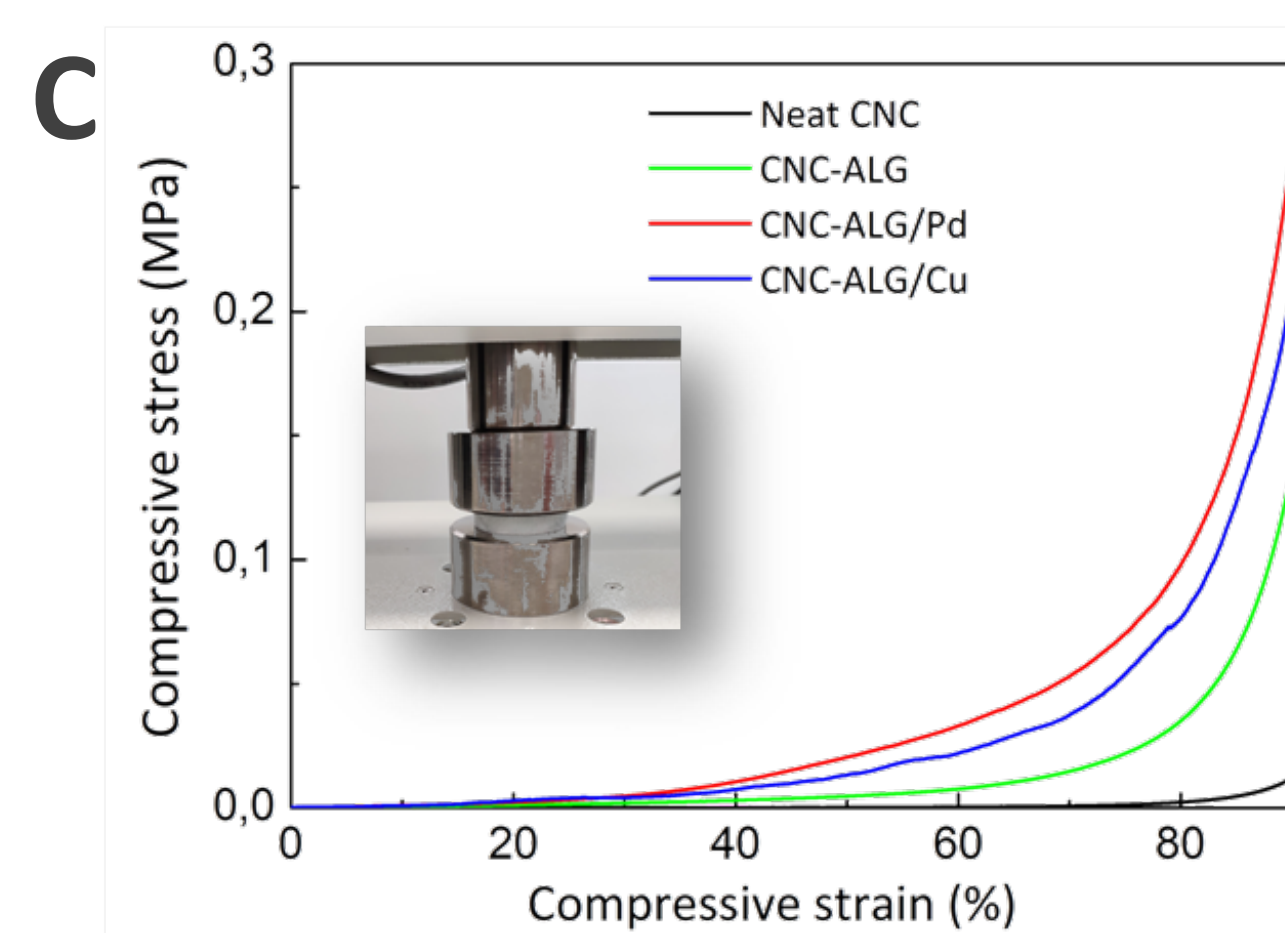


Layered structure of physically entangled CNC (a) is no altered by ZnO (b) TiO<sub>2</sub> (c) and Ag<sub>2</sub>O (d) nanoparticles addition

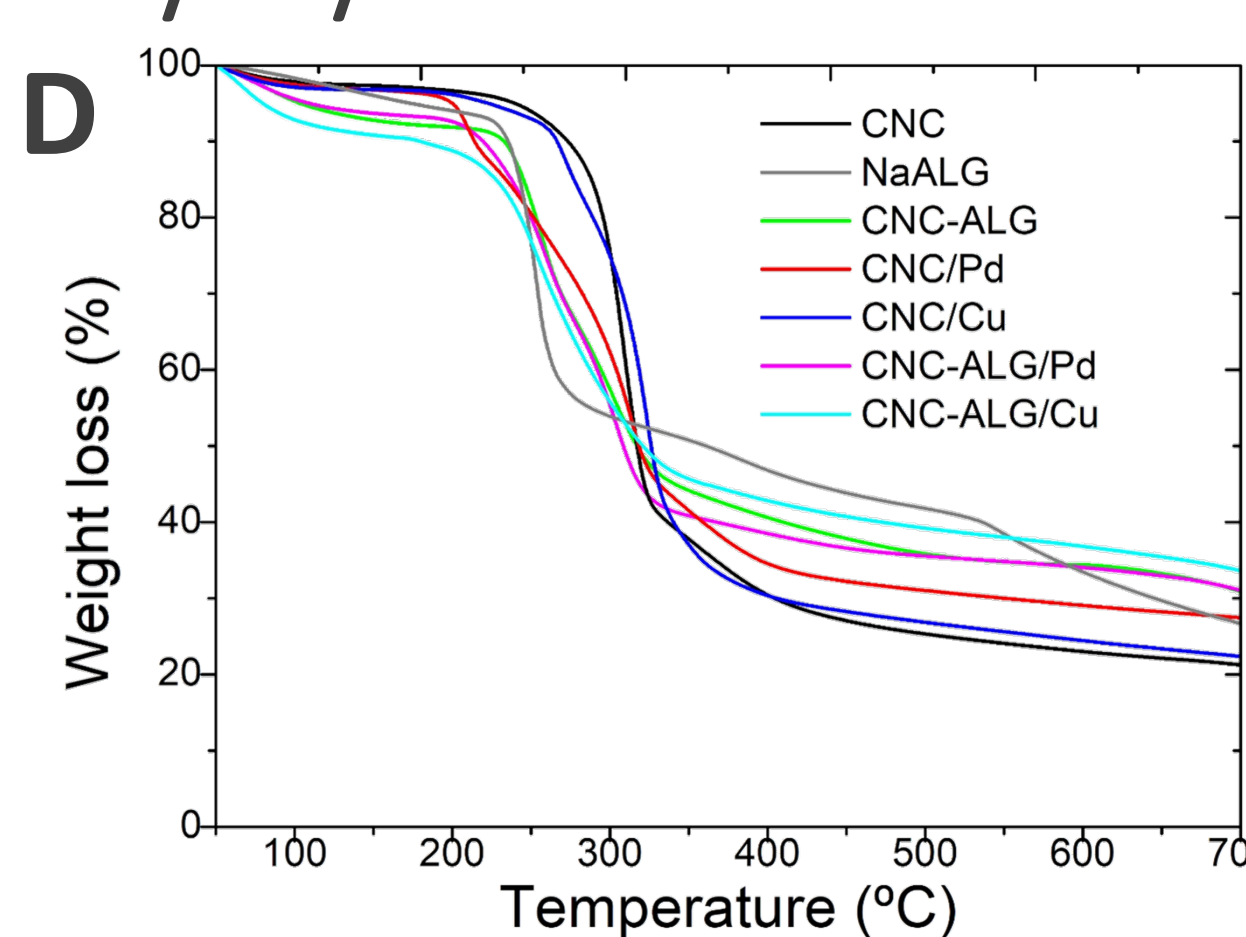


ZnO and TiO<sub>2</sub> containing films show the best disintegration behaviour at composing conditions, achieving a 70% after 90 days. CNC/Ag<sub>2</sub>O films show the worst behaviour with a 10% disintegrability

Mechanical and thermal properties

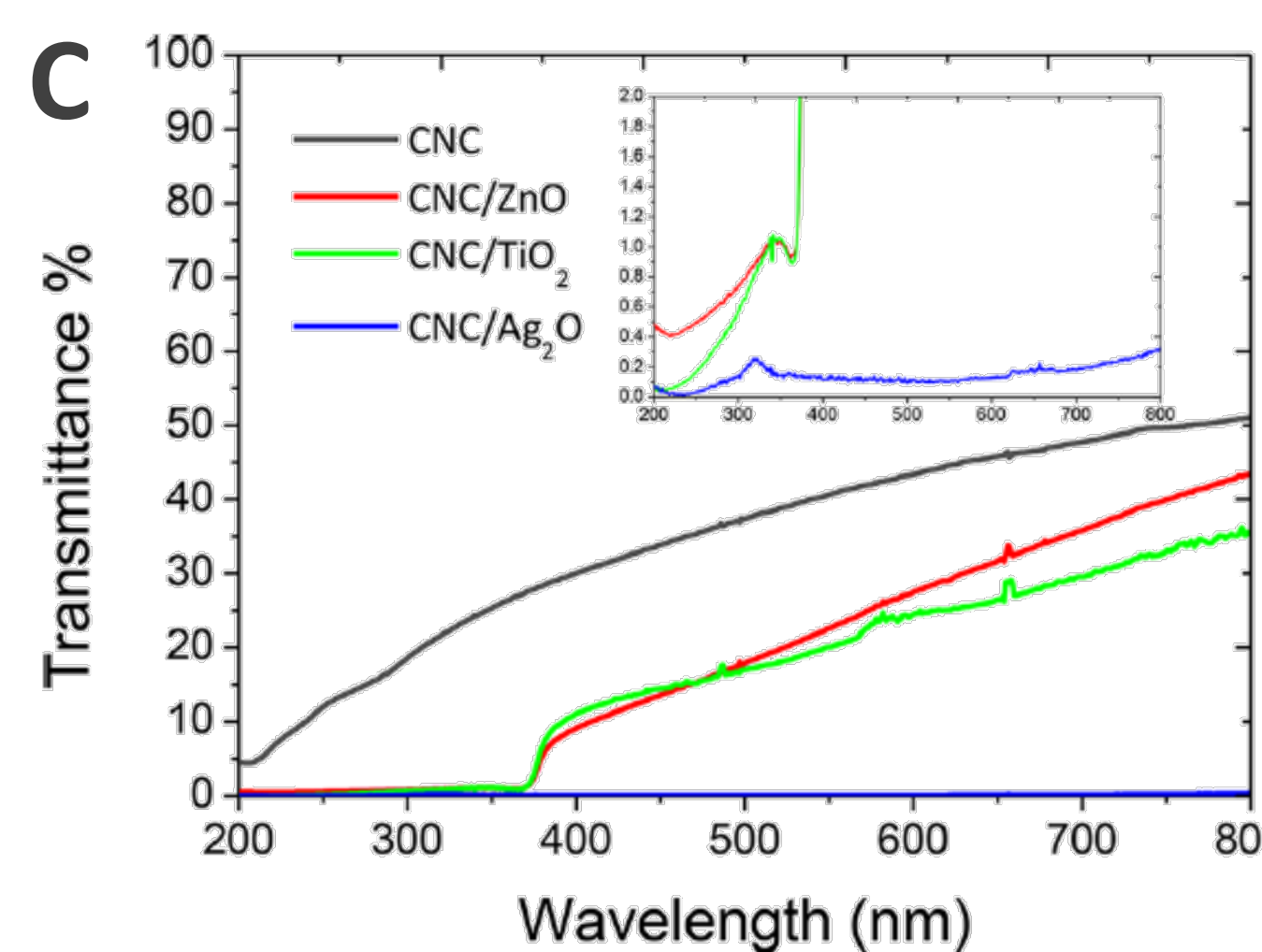


C) Aerogels crosslinked with alginate show a soft but tougher behavior, suffering total crushing but without sample fracture

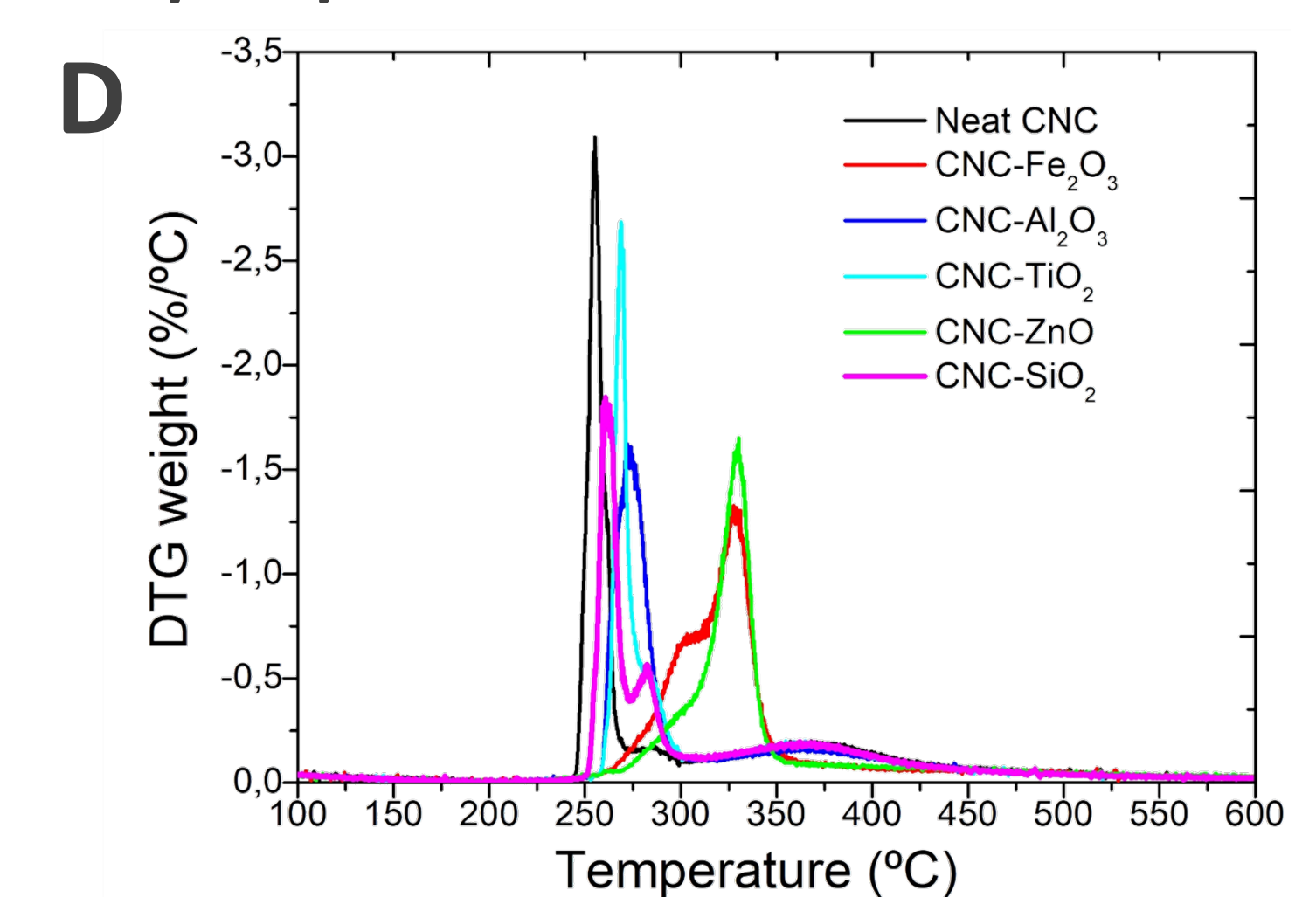


D) Neat CNC degradation curve is altered by metal and alginate addition with the presence or two degradation peaks and a lower final degradation

Optical and thermal properties



C) Metal nanoparticle addition originates UV-Visible light transmittance decrease. In the case of black coloured Ag<sub>2</sub>O oxide sample, radiation is completely absorbed, without transmittance.



D) Zn and Fe oxide nanoparticle (10% wt) containing films show a clear thermal stability improvement

CONCLUSIONS

- The generation of **entrapped palladium oxide nanoparticles** into CNC aerogels result in **hydrophobic aerogels with thermal stability improvement**.
- When CNC is **combined with alginate** via metal cation (Cu or Pd) crosslinking **mechanical performance is enhanced**.
- Metal oxide nanoparticle loaded **CNC films properties** can be **altered depending on the incorporate metal**.
- **Ti and Zn oxide** addition promote a good disintegrability behaviour, **Ag oxide changes transmittancy** and **Zn/Fe oxides** promote **thermal stability improvement**.
- The study **demonstrate the tailoring potential of cellulose nanocrystal-based structures** by the hibridation with metal oxide nanoparticles at low concentrations.

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3. Lizundia E., Puglia D., Nguyen TD, Armentano I. Cellulose nanocrystal based multifunctional nanohybrids. *Prog. Mater. Sci.* 2020,112:100668.