



CHALMERS
UNIVERSITY OF TECHNOLOGY

Solution-Spinning of a Collection of Micro- and Nanocarrier-Functionalized Polysaccharide Fibers

Downloaded from: <https://research.chalmers.se>, 2024-04-27 00:41 UTC

Citation for the original published paper (version of record):

Ulmefors, H., Yang Nilsson, T., Eriksson, V. et al (2022). Solution-Spinning of a Collection of Micro- and Nanocarrier-Functionalized Polysaccharide Fibers. *Macromolecular Materials and Engineering*, 307(8). <http://dx.doi.org/10.1002/mame.202200110>

N.B. When citing this work, cite the original published paper.

Solution-Spinning of a Collection of Micro- and Nanocarrier-Functionalized Polysaccharide Fibers

Hanna Ulmefors, Ting Yang Nilsson, Viktor Eriksson, Gustav Eriksson, Lars Evenäs, and Markus Andersson Trojer*

Continuous polysaccharide fibers and nonwovens—based on cellulose, hydroxypropyl cellulose, chitosan, or alginate—containing biopolymeric microcapsules (MC) or mesoporous silica nanoparticles (MSN) are prepared using a wet-spinning or solution blowing technique. The MCs are homogeneously distributed in the fiber matrices whereas the MSNs form discrete micron-sized aggregates as demonstrated using scanning electron-, fluorescence-, and confocal microscopy. By encapsulating the model compound pyrene, it is shown that 95% of the substance remains in the fiber during the formation process as compared to only 7% for the nonencapsulated substance. The material comprising the MC has a strong impact on the release behavior of the encapsulated pyrene as investigated using methanol extraction. MCs based on poly(L-lactic acid) prove to be practically impermeable with no pyrene released in contrast to MCs based on poly(lactic-co-glycolic acid) which allow for diffusion of pyrene through the MC and fiber as visualized using fluorescence microscopy.

1. Introduction

In order to meet the global climate challenges as established by the Glasgow agreement, there is a need to replace fossil-based raw materials with biobased and renewable equivalents.^[1] One

of the most environmentally harmful product categories is textiles, for which biobased and renewable man-made fibers still only constitute 6% of the global feedstock.^[2,3] As biobased renewable raw material replacements in general and for the production of man-made fibers in particular, polysaccharides are promising due to their inherent abundance and biodegradability. Cellulose is the most abundant biopolymer on earth and as such an almost inexhaustible source of biomass.^[4] On the fiber market, cellulose is principally represented by cotton (natural fibers) and viscose (man-made fiber) which are both suffering from water-, energy-, and chemical-intensive production processes. Chitin—the second most abundant natural polymer on Earth—is the major constituent of crustacean shells, a sizeable waste product from the food processing industry. Chitin and its

deacetylated derivative chitosan have as of yet only reached niche commercialization. Chitin is therefore considered one of the most underused polymers on Earth.^[5] Another important polysaccharide of marine origin is alginate which is a major component of the cell wall of brown algae. Alginate has mainly found use in the food industry and in niche biomedical applications.^[6] Valorization of marine biomass^[7] is one of the pillars of the blue growth initiative.^[8,9] However, to reach a future circular bioeconomy,^[1] it is pivotal to expand the application range of the polysaccharides via a wider set of properties. Historically, many chemical modification strategies have been applied to alter the properties of, and to add new functions to, polysaccharides as exemplified by celluloid, the first thermoplastic polymer^[4] or cellulose xanthogenate which is the transformation product necessary for the viscose fiber spinning process.^[10]

Besides chemical modification, encapsulating active substances with desired functions in miniature reservoirs is an efficient way to provide new and advanced properties to a material in a more general manner.^[11] The purpose of using such carrier vehicles can be narrowed down to 1) protection or to 2) controlled release of the active substances or to a combination of both.^[12] In this manner, encapsulation technology can bestow the material with a range of functions such as antimicrobial, antiviral, and antifouling properties^[13] via sustained or triggered release of bioactive substances;^[12,14,15] sensory properties and dyeing by protecting and screening chemical sensors and sensitizing colorants from the surrounding;^[16,17] means for

H. Ulmefors, T. Yang Nilsson, M. Andersson Trojer
 Department of Polymers
 Fibers and Composites, Fiber Development, RISE Research Institutes of Sweden
 Mölndal 43153, Sweden
 E-mail: markus.andersson-trojer@ri.se
 V. Eriksson, G. Eriksson, L. Evenäs
 Department of Chemistry and Chemical Engineering
 Chalmers University of Technology
 Gothenburg 41296, Sweden
 L. Evenäs
 Wallenberg Wood Science Center
 Chalmers University of Technology
 Gothenburg 41296, Sweden

The ORCID identification number(s) for the author(s) of this article can be found under <https://doi.org/10.1002/mame.202200110>

© 2022 The Authors. Macromolecular Materials and Engineering published by Wiley-VCH GmbH This is an open access article under the terms of the Creative Commons Attribution License, which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

DOI: 10.1002/mame.202200110

catalytic heterogeneous reaction by immobilization of chemical catalysts or enzymes in microreactors;^[18] odor control via odorant sorption in microcontainers, release of masking fragrances or antimicrobials;^[19] energy conservation via encapsulated phase change materials (PCM).^[20]

Most microcarrier-functionalized materials are coatings. A few examples of microcarrier-functionalized fiber materials exist in the literature of which a majority comprise microcarriers added as a finish/coating^[20,21] or embedded in a nonwoven matrix.^[22] Incorporation of microcarriers within the fiber is rare and to our knowledge limited to petroleum-based thermoplastic melt-spun fibers containing thermoset—often melamine-formaldehyde—microcapsules enveloping PCM phases.^[20]

In this paper, we present a straightforward process for the direct incorporation of a portfolio of carrier vehicles loaded with fluorophores into continuous polysaccharide fibers as well as nonwovens via regular solution spinning or solution blowing respectively. This is to our knowledge the first publication describing the incorporation of (thermo)sensitive carrier vehicles into fiber materials. The carrier vehicles are in this study either biopolymer-based microcapsules (MC) or mesoporous silica nanoparticles (MSN) for the encapsulation of hydrophobic or hydrophilic actives respectively. In contrast to the PCM example mentioned above, our spinning process allows for a wide range of temperature-sensitive and biodegradable biopolymer-based MCs to be incorporated. The polysaccharides constituting the fiber matrix are either cellulose, the cellulose derivative hydroxypropyl cellulose (HPC), alginate, or the chitin derivative chitosan. The cellulose fibers are water-insoluble whereas chitosan, alginate, and post-crosslinked HPC^[23] form hydrogelling fiber materials. Most focus in this work is devoted to cellulosic fibers, which have the most widespread use and potential of the investigated polysaccharides, containing aliphatic polyester-based poly(lactic-co-glycolic acid) (PLGA) or poly(L-lactic acid) (PLLA) MCs. These are among the most used and studied biobased polymer materials.

2. Results and Discussion

Below, we start by describing the fiber formation process and discussing the conditions for producing MC- and MSN-functionalized polysaccharide filaments and nonwovens. Thereafter, the fiber structure for a collection of MC- and MSN-functionalized polysaccharide filaments and nonwovens are displayed. The fiber/particle morphology has been studied using a range of microscopy methods; electron microscopy (SEM, TEM), optical microscopy (bright field, fluorescence, cross-polarization), and confocal microscopy. Finally, a selection of illustrative results, which demonstrate the advantage of encapsulation for fiber functionalization in terms of the fate and distribution of the model actives (fluorophores) in the multicompartment fiber materials—which have been studied using spectroscopy (UV-vis spectrophotometry and NMR spectroscopy)—are presented.

2.1. Formation of Carrier Vehicle-Containing Fiber Materials

The polysaccharide fibers—including both continuous wet-spun filaments^[24] and solution-blown nonwovens^[23]—containing

MCs or MSNs were formed using a bench-scale multifilament solution spinning or solution blowing equipment (see **Figure 1**). In more detail, the MCs or MSNs—prepared using the internal phase separation method^[11] or a microemulsion-templated condensation method^[25]—were dispersed in the polysaccharide dope solution (at a concentration of 5 wt% of the total dry weight unless otherwise stated). The dope solution was subsequently extruded and precipitated in a coagulation bath or by heating/evaporation. The HPC, alginate, and chitosan fibers were spun from aqueous-based dope solutions whereas the cellulose fibers were spun from ionic liquid-based dope solutions with or without cosolvents (see the Supporting Information).

The MC and MSN particles were formulated to contain fluorophores (**Figure 1**). The purpose of using fluorophores is multi-fold. They are easily detected using standard UV-vis or fluorescence micro-/spectroscopy and serve as model compounds for low molecular weight actives with similar physicochemical properties. In addition, the fluorophores are used to elucidate the particle morphology (in aqueous suspension and in the polysaccharide fiber).

2.1.1. Influence of the Process Parameters

Several process parameters, which are summarized in **Figure 1**, must be considered for the successful incorporation of loaded MSNs and MCs into the polysaccharide fibers. The spinnability of the dope solution depends on its viscoelastic properties which in turn depends on temperature, the concentration of the fiber-forming polysaccharide, and of the incorporated MCs or MSNs. This is of particular importance for the cellulose-based fibers which are spun from the ionic liquid ethylmethylimidazolium chloride (EMIMAc). EMIMAc dissolves a range of fluorophores and semihydrophobic polymers, and the spinning conditions often require elevated processing temperature (see below). For HPC, alginate, and chitosan, the viscoelastic properties of the dope are dominated by the polysaccharide, at least within the MC/MSN concentration interval (2–20%) analyzed in this study. However, for MC-functionalized cellulose fibers, higher concentrations of capsules (>10%) resulted in dope gelation (see **Figure S4** in the Supporting Information) due to the addition of the residual antisolvent water which is difficult to completely avoid from the concentrated MC suspension (containing approximately 50 wt% water, **3** in **Figure 1**).

Regarding the effect of temperature, regenerated cellulose fibers are usually prepared at elevated temperature (dope dissolution and solution blowing at 70 °C). MCs with high glass transition temperature such as PMMA can tolerate such conditions. However, for more temperature-sensitive MCs such as PLGA, the high process temperature results in fragmentation (see **Figure S3** in the Supporting Information). Elevated temperatures can be circumvented by either reducing the cellulose concentration or by using a cellulose grade with a lower average molecular weight—such as microcrystalline cellulose ($M_w \approx 78 \text{ kg mol}^{-1}$) instead of dissolving pulp (α -cellulose, $M_w = 324 \text{ kg mol}^{-1}$)—for which room temperature processing is feasible. The viscosity of the dope can also be adjusted by adding highly polar aprotic cosolvents such as dimethylsulfoxide or methylimidazole^[10] which is also important for reducing die-swelling during regular

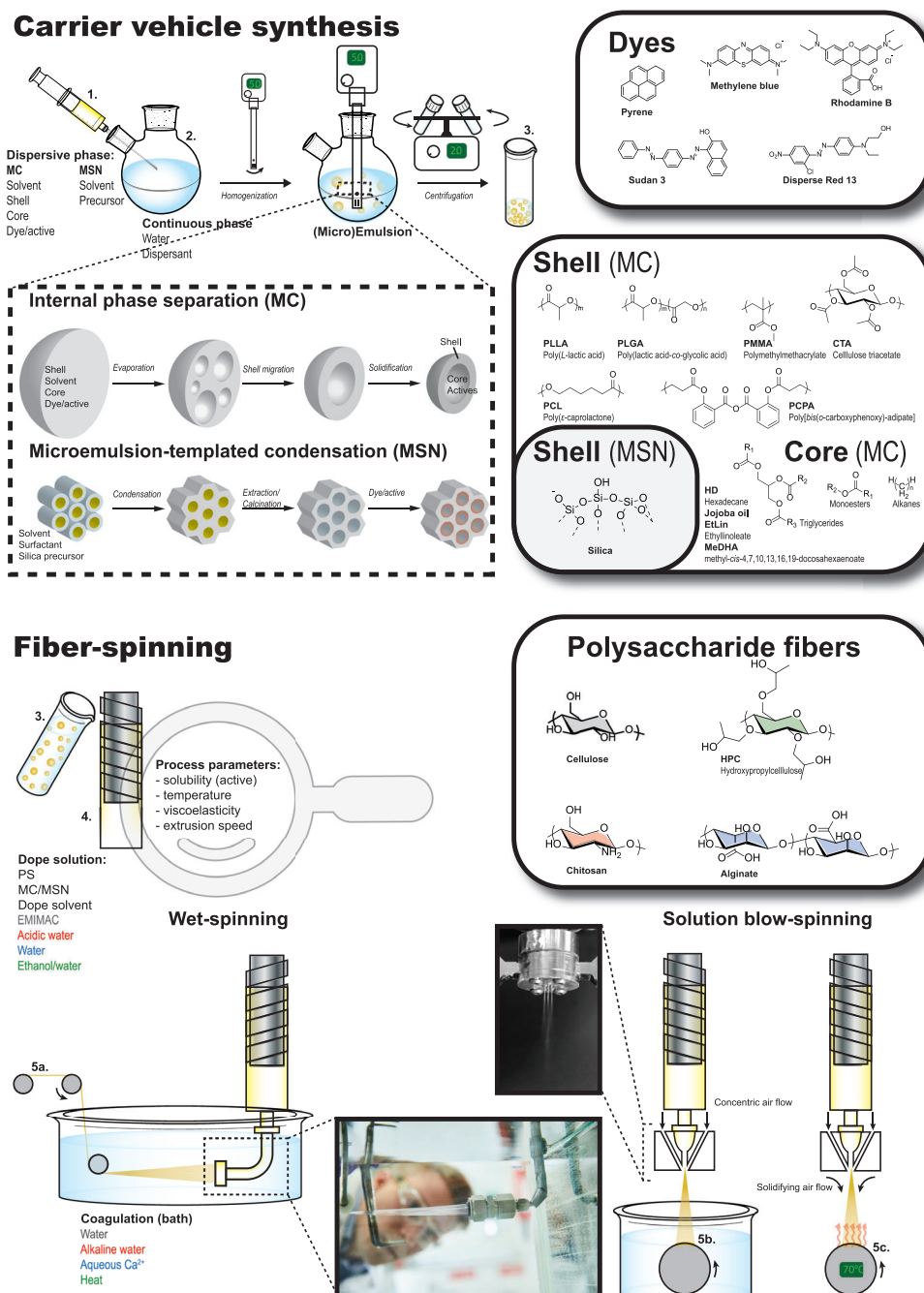


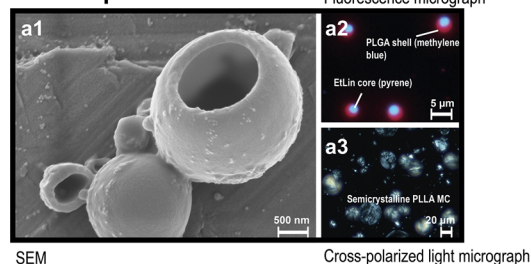
Figure 1. Schematic overview of the carrier vehicle syntheses for MC and MSN production (1.-3.), and the fiber-spinning process from dope preparation to wet-spinning or solution blowing (4.-5.). The portfolio of carrier vehicle materials (shell and core), dyes, and fiber-forming polysaccharides are also summarized. The polysaccharides have been color-coded to indicate the dope solution and coagulation bath composition.

wet-spinning. This brings the discussion to the effect of the dope solvent. MSNs remain perfectly intact in EMIMAC/cosolvent dope solutions (see Figure 2f1,f2) but dissolve in the acidic dope of chitosan (see Figure S5 in the Supporting Information). In contrast, the MCs are perfectly stable in all aqueous-based dope and coagulation bath systems but disintegrate in most EMIMAC/cosolvent dopes containing high amounts of cosolvent (see Figures S7 and S8 in the Supporting Information). It is

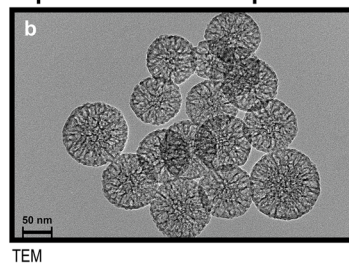
possible to find specific MC-cosolvent pairs, such as PCL-based MCs in EMIMAC/propylene carbonate-based dope solvents, which enables the formation of cellulose fiber-containing intact MCs (data not shown). However, a more general solution for cellulose fiber formation using high concentrations of dissolving pulp is air gap-spinning which, similar to solution blowing, extrudes the filaments into the air prior to precipitation in the coagulation bath.

Carrier vehicles

Microcapsules

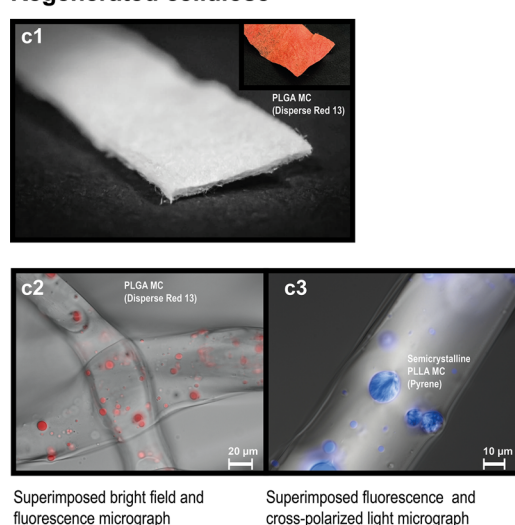


Mesoporous silica nanoparticles

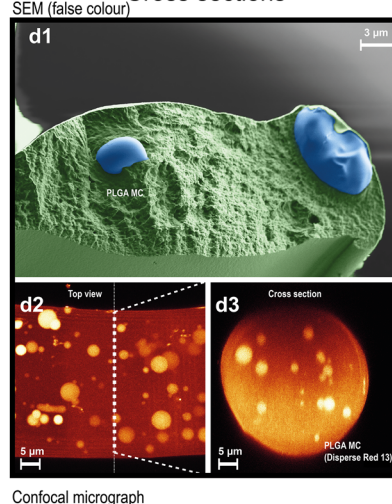


Filaments and nonwovens

Regenerated cellulose

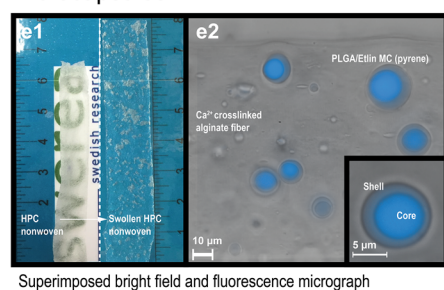


Cross sections



Hydrogel-forming fibers

Microcapsules



Mesoporous silica nanoparticles

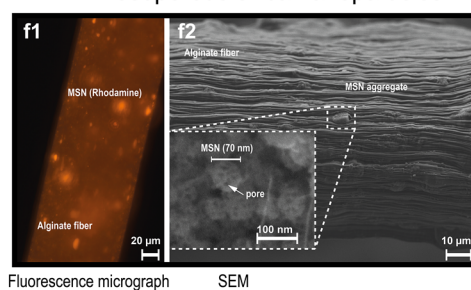


Figure 2. Collage displaying a selection of carrier vehicles a,b) and fiber materials (filaments and nonwovens, c–f) containing incorporated carrier vehicles. SEM image of PLGA MCs (a1). Fluorescence micrograph of stained PLGA/EtLin MCs (a2). Cross-polarized light micrograph of PLLA MCs (a3). TEM image of MSN particles (b). In c,d), cellulosic filaments and nonwovens containing MCs are shown. In (c1), a cellulose nonwoven mesh is shown. The inset displays the same material containing PLGA-encapsulated Disperse Red 13 which is also magnified in (c2) displaying the fiber structure and the stained MCs. Superimposed fluorescence and cross-polarized light micrograph of PLLA MCs in cellulose fiber (c3). d) Fiber cross-sections of cellulose materials from (c1) and (c2) are shown using SEM (d1) and confocal microscopy (d2 and d3). The top view of the fiber is shown in (d2) and the dashed white line indicates the cross-section in (d3). Hydrogel-forming fibers are shown in (e) and (f). In (e1), the reversible gelling of a HPC nonwoven is shown. In (e2), a superimposed bright field and fluorescence micrograph image show PLGA/EtLin MCs in alginate fibers. In (f1), alginate fibers containing MSN-encapsulated RhB is shown. In (f2), a SEM image of the fiber from (f1) is shown with the inset displaying a self-assembled MSN particle.

2.2. Fiber Morphology and Carrier Vehicle Distribution

In Figure 2, a collection of carrier vehicle-polysaccharide fiber combinations is displayed. As seen, the MCs are in the micron range with a relatively narrow size distribution (Figure 2a), whereas the mesoporous MSNs are approximately 100 nm (Figure 2b in which the pore structure is visible). For microcapsules of core-shell morphology, the hollow interior can be visualized using SEM. In Figure 2a1, a microcapsule has been gently heated by increasing the acceleration voltage, which combined with the vacuum caused the thin shell to collapse into the large uniform cavity. Moreover, using fluorophores of different hydrophobicity, it is possible to selectively stain the MC shell (PLGA stained with methylene blue) and the core (ethyl linoleate, EtLin, stained with pyrene) as seen in Figure 2a2. However, in the remaining micrographs, only one fluorophore has been used in order to qualitatively assess its distribution in the core, the shell, and the fiber matrix. Let us first consider MCs incorporated into fibers. Cross-sections of the fibers (SEM and confocal microscopy, see Figure 2d and Figure S9 in the Supporting Information) show that the MCs are randomly distributed inside the fiber matrix. In addition, the crystallinity of semicrystalline MCs, as characterized using cross-polarized light microscopy (Figure 2a3), is maintained in the fiber. This is revealed in Figure 2c3 using cross-polarized light where the PLLA-based MCs display an optical rotation different from that of the birefringent cellulose matrix.^[24] Figure 2e2 displays MCs of core-shell morphology, which is clearly visible in the inset with pyrene residing in the EtLin core, dispersed in alginate fibers. MCs of such morphology are more sensitive to the extrusion rate than MCs consisting of a homogenous polymer matrix (see Figure S6 in the Supporting information). At higher extrusion rates, the spherical core-shell particles become slightly ellipsoid, yet with intact structural integrity.

Regarding MSN-functionalized fibers, the nanoparticles form discrete micron-sized aggregates in the presence of the polysaccharide chains which are stretched in the fiber direction. This is a phenomenon that has been previously observed for nonporous silica nanoparticles in cellulose fibers and is strongly dependent on the surface chemistry of the particles.^[24] The particle aggregates are however randomly distributed in the fiber matrix (see Figure 2f in which the individual MSNs and the pores are clearly visible in the inset of Figure 2f2) and the model compound rhodamine B (RhB) remains adsorbed in the MSNs (see Figure 2f1).

2.3. Loading Efficiency

The carrier vehicles provide protection of the actives during the spinning process, which is demonstrated using the model compounds pyrene and RhB in MCs and MSNs respectively. Among the different polysaccharide fiber systems investigated in this work, this is particularly important for cellulose fiber formation. As the cellulose dope solution enters the coagulation bath, there is a net flow of dope solvent (ionic liquid) from the precipitating fiber to the surrounding medium (see Figure 3).^[26] This mass transport may result in a depletion of actives in the fiber. For pyrene simply dissolved in the cellulosic dope solution, only 7% remains in the final fiber as established using UV-vis spec-

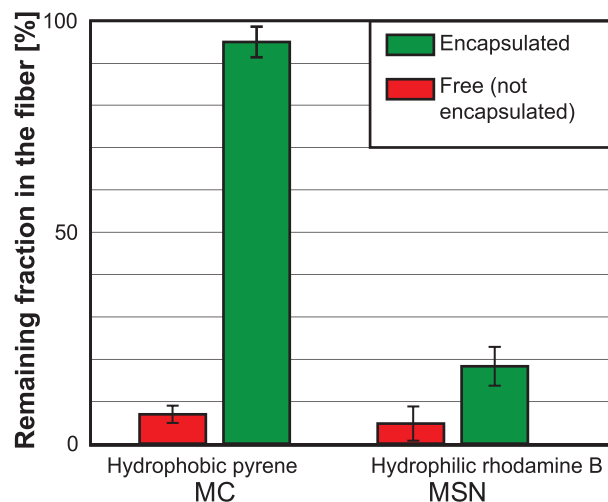


Figure 3. Fraction of encapsulated (MC or MSN) and free active (pyrene or RhB) remaining in the cellulose fiber.

troscopy despite the high hydrophobicity of pyrene. Note that the final pyrene concentration in the coagulation bath is above its saturation concentration in water which suggests that the loss is related to the mass transport of dope solvent during the coagulation. In contrast, for pyrene encapsulated in PLGA MCs, 95% of the pyrene remains in the fiber, as based on the amount encapsulated in 1. in Figure 1. In addition, quantitative NMR analysis reveals that the ratio of pyrene/MC remains constant in the aqueous suspension, in the dope, and in the solid fiber.

Regarding RhB, the incorporation yields are significantly lower compared to those for pyrene. This is most likely a result of 1) the high water solubility of the dye and 2) the mode of encapsulation. It is important to note that RhB is physically adsorbed in the pores of MSN and hence more exposed the dope solution and extraction solvents as compared to pyrene which is dispersed or dissolved in the MC shell matrix or oil core respectively. Nevertheless, the use of MSNs improves the incorporation yield of RhB into the fibers more than threefold.

2.4. Effect of the MC Material on the Release

We have below illustrated, using a simple experiment, how the proper choice of carrier vehicle can be used for either protection or controlled release of an active. Note that the function of the carrier vehicle depends on its barrier properties. For protection, the shell should be impermeable. For controlled release, the shell should be permeable or be rendered permeable by a triggerable event in order to allow for diffusion through the shell barrier. In Figure 4, fluorescence micrographs of two types of cellulose fibers containing pyrene encapsulated in either semicrystalline PLLA or amorphous PLGA are displayed. As a droplet of methanol (MeOH), which is a good solvent for pyrene and penetrates the cellulose matrix efficiently, is positioned on the fiber mesh, it can be observed that pyrene is extracted from the PLGA reservoirs into the fiber matrix. It should be noted that the concentration of pyrene is still higher inside the MCs than in the fiber matrix. Yet, this is still in sharp contrast to the extraction

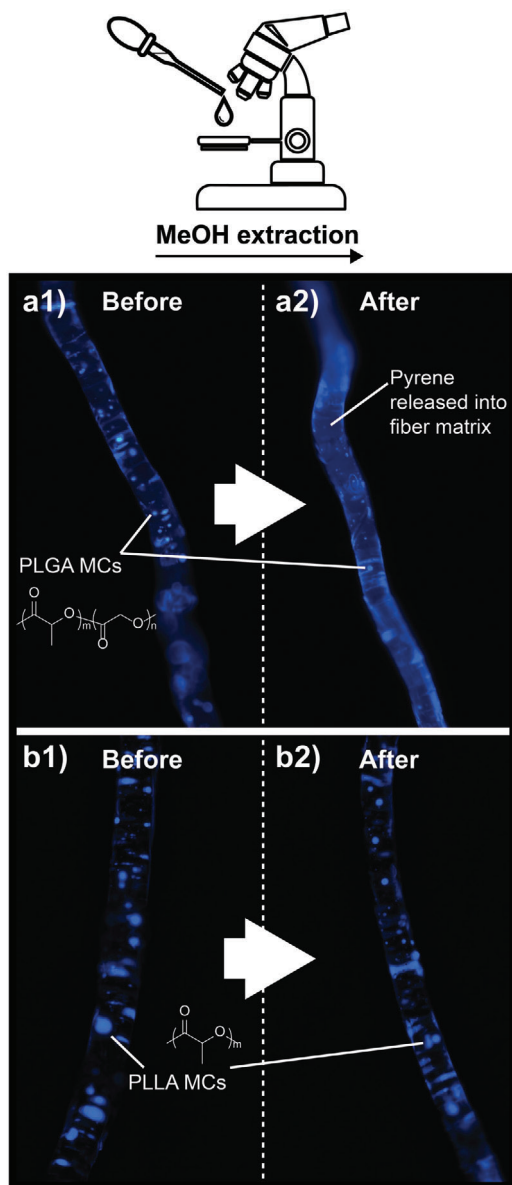


Figure 4. MeOH extraction of pyrene from a) PLGA MCs or b) PLLA MCs in fibers. The left images (1) display the pristine fibers and in the right images (2) a drop of MeOH has been placed on the fibers to follow the extraction process in situ under the fluorescence microscope.

results for PLLA-based MCs from which no pyrene is extracted. This effect is ascribed to a very low pyrene diffusivity in PLLA due to large impermeable crystalline domains of the polymer (see Figure 2).

3. Conclusion

A wet-spinning technique, which enables a multitude of sensitive carrier vehicles to be incorporated into fibers of the important polysaccharides cellulose and hydroxypropyl cellulose (HPC), and the marine polysaccharides alginate and chitosan, is presented. The biopolymer-based microcapsules (MC) can be incorporated into all investigated polysaccharide fibers as long

as aprotic solvents and elevated temperatures are avoided during cellulose wet-spinning. The mesoporous silica nanoparticles (MSN) can be incorporated into HPC, alginate and all tested cellulose fibers but dissolve in the acidic chitosan dope. The use of carrier vehicles is clearly a resource-efficient method for incorporating actives into wet-spun fibers, in particular when using MCs for the incorporation of hydrophobic dyes. The MSN loading efficiency and control of the dispersibility/aggregation in the fiber could however be improved in order to reach a more mature technology readiness level. The proper choice of carrier vehicle can bestow the fiber with tailored functions. As demonstrated, MCs based on semicrystalline PLLA can be used for the protection of actives, which are not intended to be released, e.g., molecular sensors. On the other hand, PLGA is well suited for sustained release applications as will be the topic of a separate publication.^[27]

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

Acknowledgements

The Swedish Research Council FORMAS (2018–02284 and 2021–02642) and Vinnova (2017–04693 and 2021–01611) are acknowledged for funding. Dr. Mats Hulander and Dr. Annika Krona are acknowledged for SEM and confocal micrographs respectively.

After initial online publication, the affiliation Wallenberg Wood Science Center was affiliated to author L.E. on May 3, 2022, after being previously incorrectly affiliated to author M.A.T.

Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available in the supplementary material of this article.

Keywords

core-shell particles, filaments, nonwovens, polysaccharides, solution blown

Received: February 15, 2022

Revised: March 17, 2022

Published online:

- [1] P. Stegmann, M. Londo, M. Junginger, *Resour. Conserv. Recycl.* **2020**, *6*, 100029.
- [2] K. Niinimäki, G. Peters, H. Dahlbo, P. Perry, T. Rissanen, A. Gwilt, *Nat. Rev. Earth Environ.* **2020**, *1*, 189.
- [3] G. Sandin, G. M. Peters, *J. Cleaner Prod.* **2018**, *184*, 353.
- [4] D. Klemm, B. Heublein, H. - P. Fink, A. Bohn, *Angew. Chem., Int. Ed.* **2005**, *44*, 3358.
- [5] O. Nechyporchuk, T. Yang Nilsson, H. Ulmefors, T. Köhnke, *ACS Appl. Polym. Mater.* **2020**, *2*, 3867.

- [6] I. A. Brownlee, C. J. Seal, M. Wilcox, P. W. Dettmar, J. P. Pearson, in *Alginates: Biology and Applications* (Ed: B. H. A. Rehm), Springer, Berlin **2009**, p. 211.
- [7] C. Xu, M. Nasrollahzadeh, M. Selva, Z. Issaabadi, R. Luque, *Chem. Soc. Rev.* **2019**, *48*, 4791.
- [8] R. Carrasco-Reinado, A. Escobar, C. Carrera, P. Guarnizo, R. A. Vallejo, F. J. Fernández-Acero, *LWT* **2019**, *114*, 108385.
- [9] L. Mulazzani, G. Malorgio, *Mar. Policy* **2017**, *85*, 17.
- [10] C. Olsson, G. Westman, in *Cellulose: Fundamental Aspects* (Eds: T. van de Ven, L. Godbout), IntechOpen, London **2013**, p. 143.
- [11] M. Andersson Trojer, L. Nordstierna, J. Bergek, H. Blanck, K. Holmberg, M. Nydén, *Adv. Colloid Interface Sci.* **2015**, *222*, 18.
- [12] M. Andersson Trojer, L. Nordstierna, M. Nordin, M. Nydén, K. Holmberg, *Phys. Chem. Chem. Phys.* **2013**, *15*, 17727.
- [13] J. Bergek, M. Andersson Trojer, H. Uhr, L. Nordstierna, *J. Controlled Release* **2016**, *225*, 31.
- [14] V. Eriksson, M. Andersson Trojer, S. Vavra, M. Hulander, L. Nordstierna, *J. Colloid Interface Sci.* **2020**, *579*, 645.
- [15] A. P. Esser-Kahn, S. A. Odom, N. R. Sottos, S. R. White, J. S. Moore, *Macromolecules* **2011**, *44*, 5539.
- [16] S. Chinnayelka, M. J. Mcshane, *Anal. Chem.* **2005**, *77*, 5501.
- [17] L. I. Kazakova, L. I. Shabarchina, G. B. Sukhorukov, *Phys. Chem. Chem. Phys.* **2011**, *13*, 11110.
- [18] S.-I. Matsuura, T. Yokoyama, R. Ishii, T. Itoh, E. Tomon, S. Hamakawa, T. Tsunoda, F. Mizukami, H. Nanbu, T.-A. Hanaoka, *Chem. Commun.* **2012**, *48*, 7058.
- [19] N. Paret, A. Trachsel, D. L. Berthier, A. Herrmann, *Angew. Chem., Int. Ed.* **2015**, *54*, 2275.
- [20] Y. Yan, in *Advances in Technical Nonwovens* (Ed: G. Kellie), Woodhead Publishing, Sawston, UK **2016**, p. 19.
- [21] M. Martí, M. Lis, J. A. Navarro, R. Ramírez, L. Coderch, J. Valldeperas, J. L. Parra, in *Medical and Healthcare Textiles* (Eds: S. C. Anand, J. F. Kennedy, M. Mirafteb, S. Rajendran), Woodhead Publishing, Sawston, UK **2010**, p. 509.
- [22] M. Grandcolas, T. O. Sunde, M. Pilz, C. Simon, R. Schmid, H. Johnsen, *Int. J. Eng. Res. Appl.* **2017**, *07*, 100.
- [23] T. Yang Nilsson, M. Andersson Trojer, *Soft Matter* **2020**, *16*, 6850.
- [24] M. Andersson Trojer, C. Olsson, J. Bengtsson, A. Hedlund, R. Bordes, *J. Colloid Interface Sci.* **2019**, *553*, 167.
- [25] H. Gustafsson, S. Isaksson, A. Altskär, K. Holmberg, *J. Colloid Interface Sci.* **2016**, *467*, 253.
- [26] A. Hedlund, T. Köhnke, H. Theliander, *Macromolecules* **2017**, *50*, 8707.
- [27] V. Eriksson, J. Mistral, T. Y. Nilsson, M. A. Trojer, L. Evenäs, *Microencapsulation for controlled release of model active substances from cellulose nonwovens*, **2022**.