Microplastic-free microcapsules using interfacial self-assembly of bis-urea molecules

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UNIVERSITY OF BIRMINGHAM

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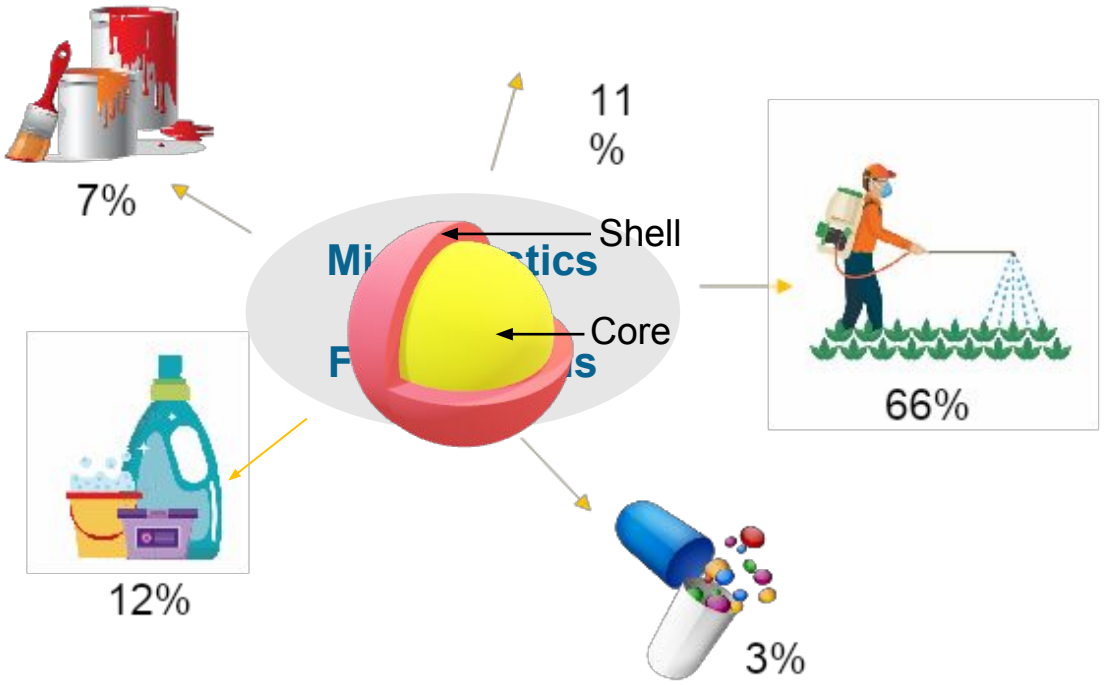
INTEGRATING
Delivery Science
ACROSS DISCIPLINES

Background



Microcapsules are widely used in these commercial formulations for:

- Protection, increasing shelf-life
- Easier handling
- **Controlled release**

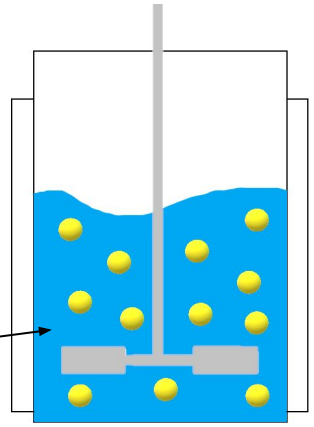
Capsule shell from **synthetic polymers**, highly crosslinked, non-biodegradable – **MICROPLASTIC**

Solution: Using **small molecules**, self-assembled with **non-covalent** hydrogen bonds to form the shell

Sector-wise annual microplastic waste from formulations in the EEA in 2017 (**ca. 36,000 tonnes microplastic** released in total)

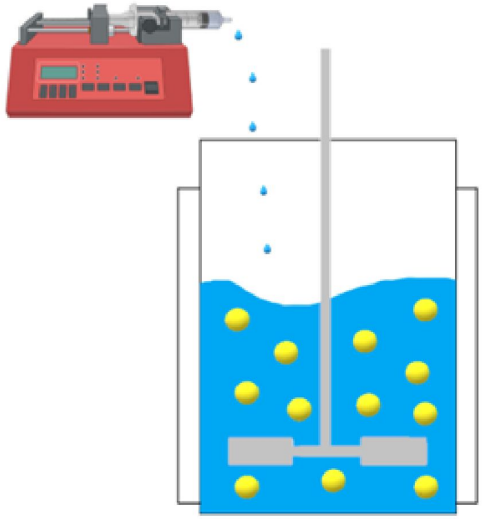
Strategy

Step 1
- Stable oil-in-water emulsion

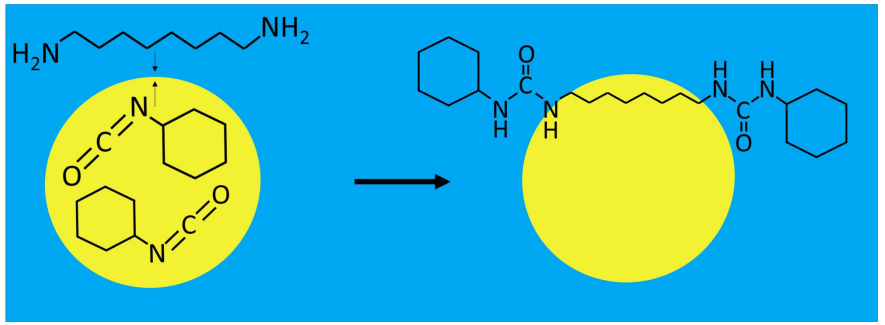


- Oil Phase
  - Active Ingredient
  - Isocyanate

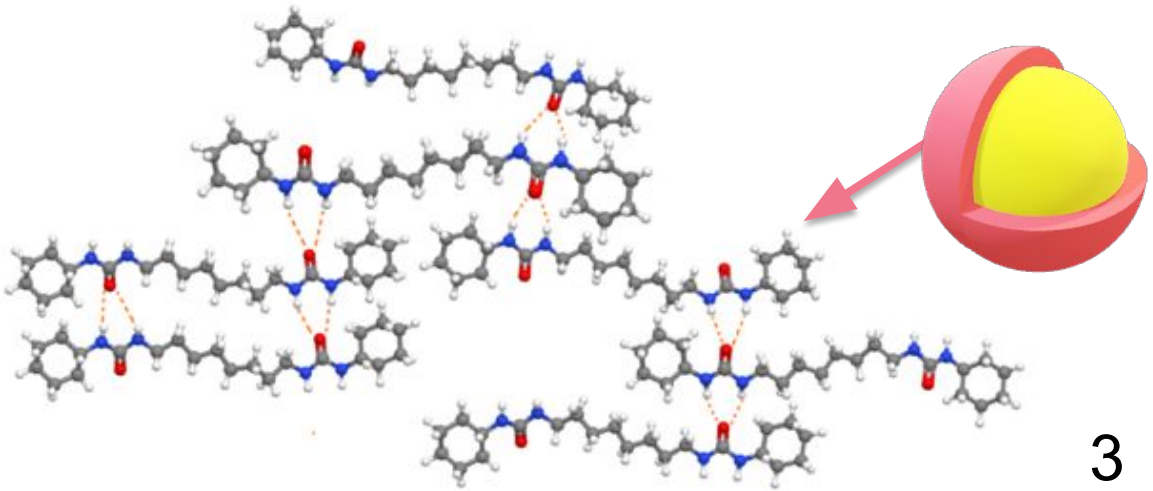
Step 2
- Amine addition



Step 3
- Reaction and bis-urea formation at the interface



Step 4
- Self-assembly at the interface to form the shell



***In-situ* generation and interfacial self-assembly of bis-urea molecules**

Observations

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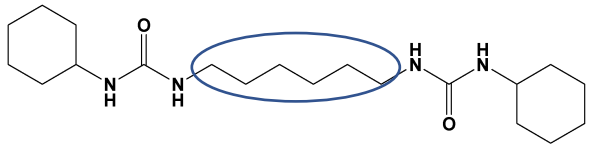
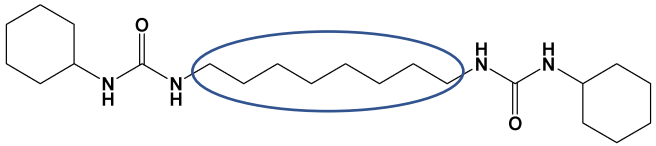
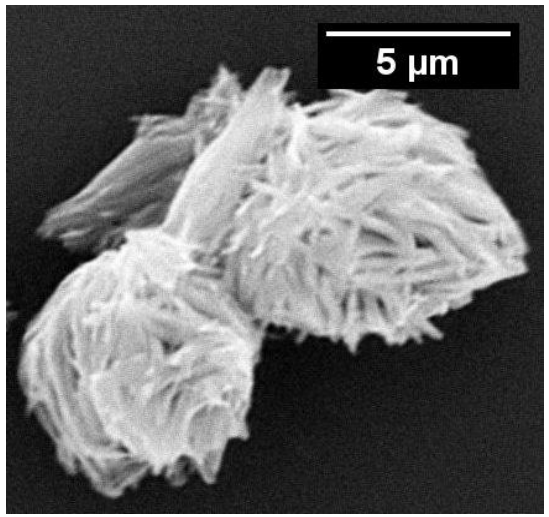
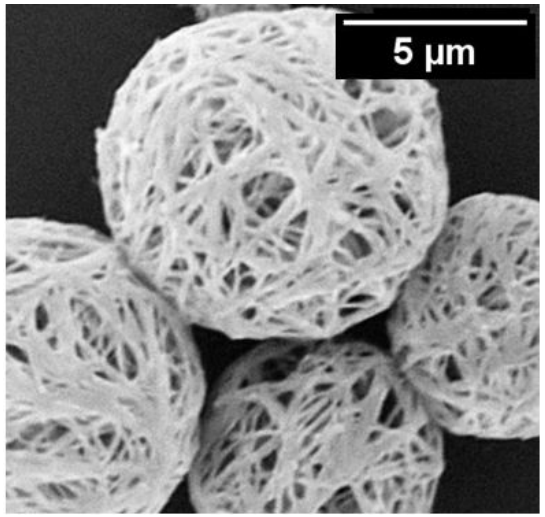
- **Morphology**

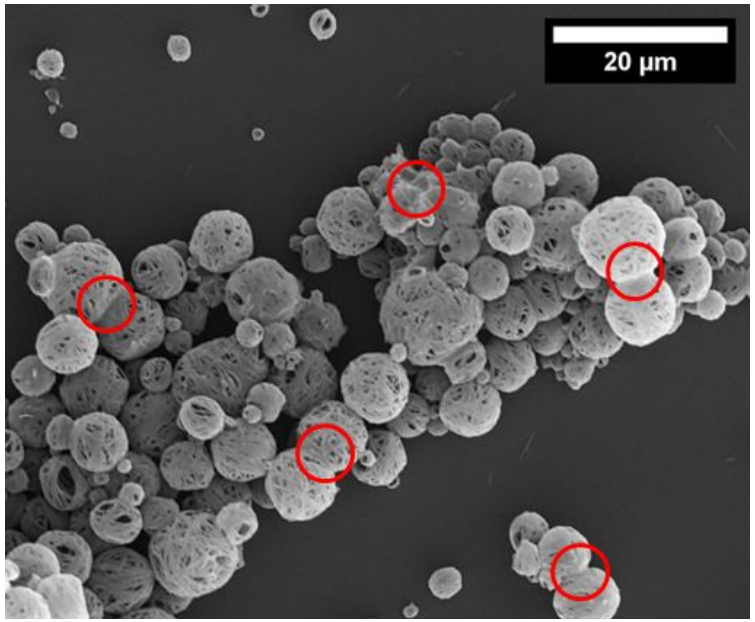


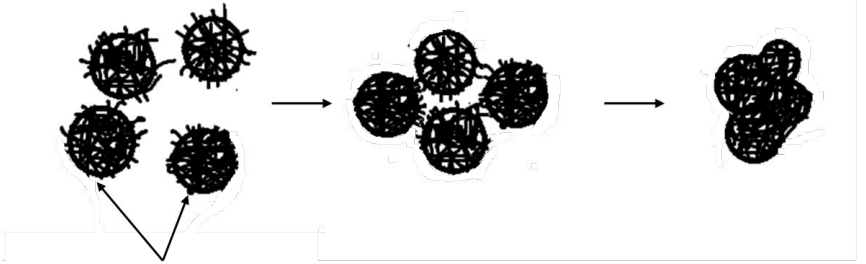






- **Aggregation**

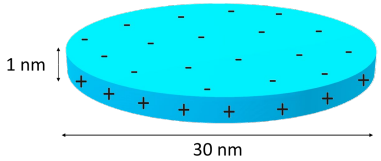


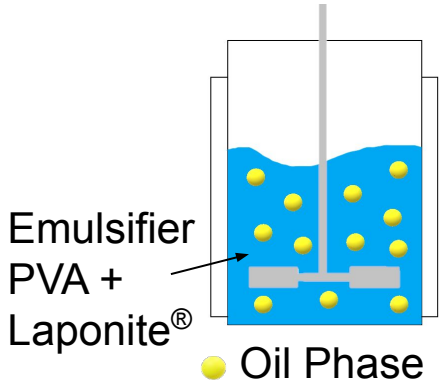


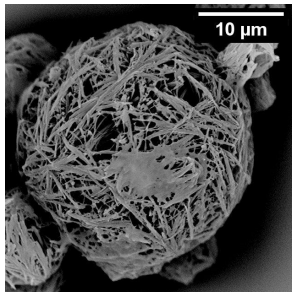
Reduce aggregation and porosity

Improvements

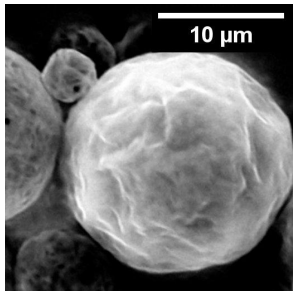
- **Laponite® nanodiscs**







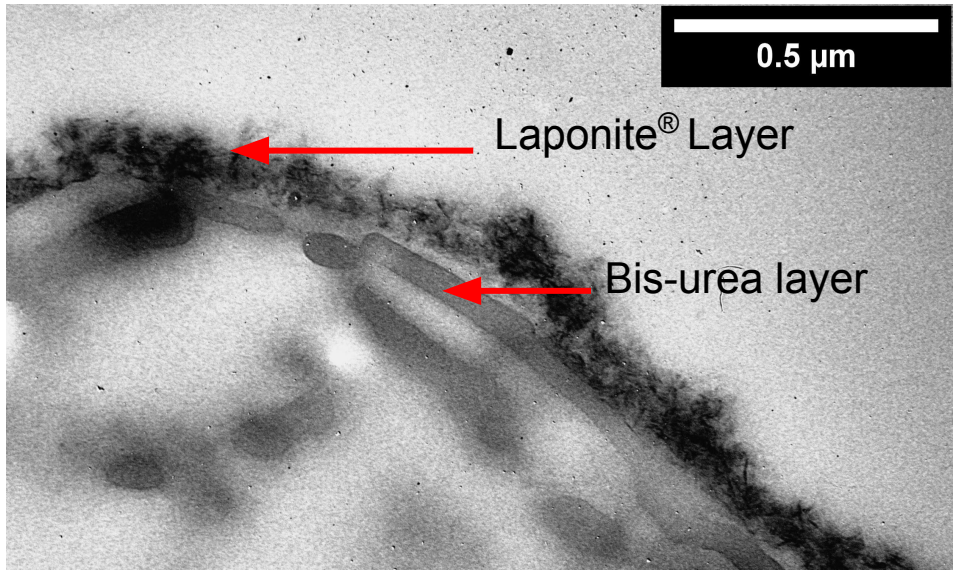
Without Laponite®



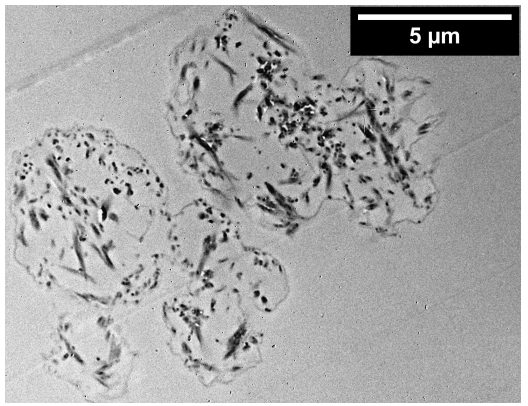
With Laponite®

Volume (%)

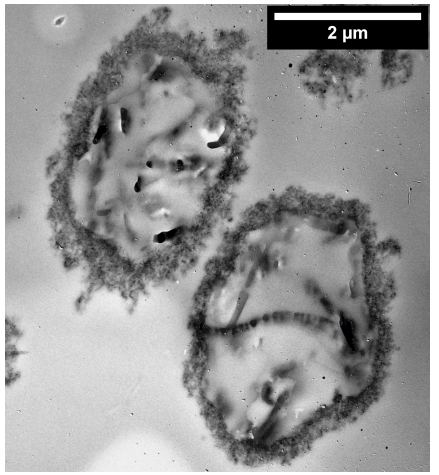
Particle Size (µm)



- **TEM Cross-section**



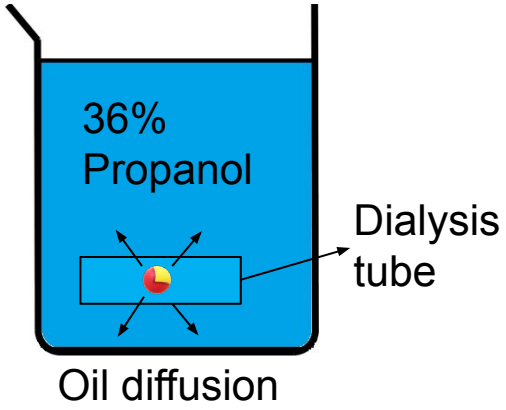
Without Laponite®



With Laponite®

Release Study

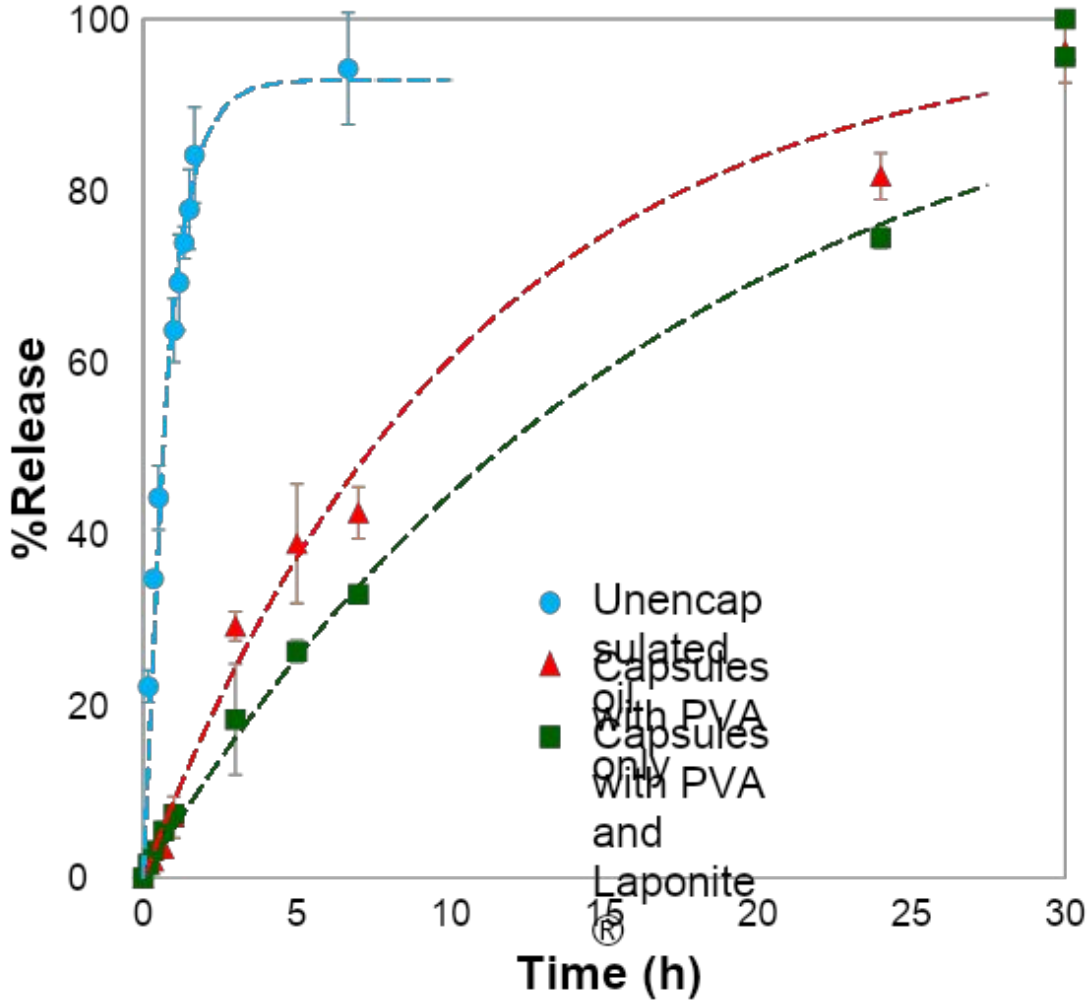
- Accelerated release test in 36% aq. propanol (v/v)



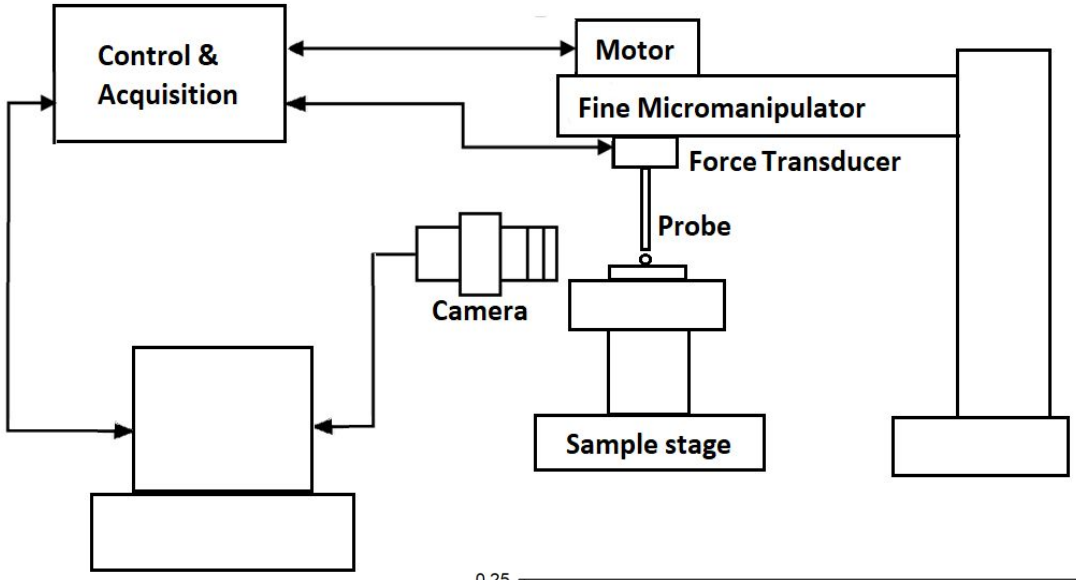
$$y = a\left(1 - e^{-\frac{t}{\tau}}\right)$$

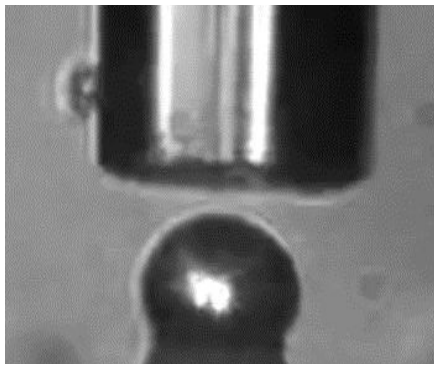
y: cumulative release (weight fraction)
t: time
a: dimensionless constant
τ: characteristic release time

	τ (min)
Unencapsulated oil	47
Capsules made using PVA only	634
Capsules made using PVA and Laponite ®	1017

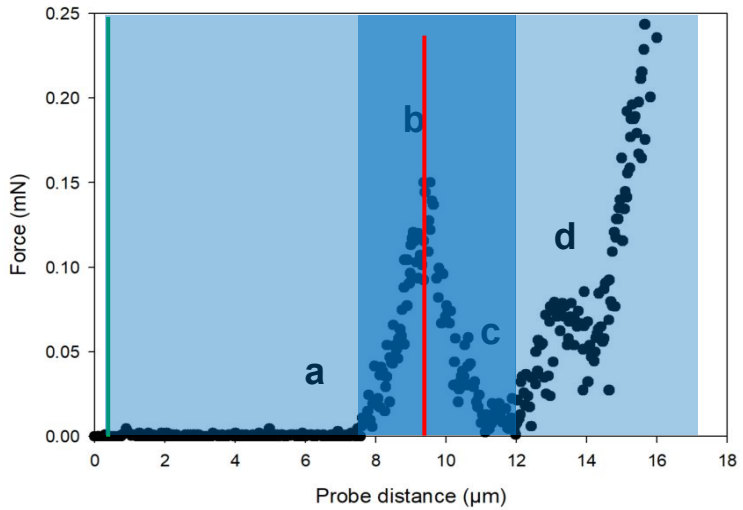


Mechanical Strength





Capsule dia = 20 µm



	Rupture tension (µN/µm)
Capsules made using PVA only	**7 ± 1**
Capsules made using PVA and Laponite ®	**23 ± 2**

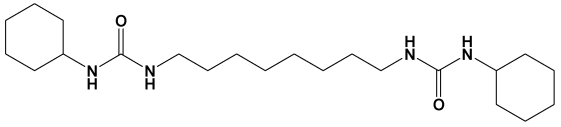
$$\text{Rupture tension} = \frac{F_r}{d}$$

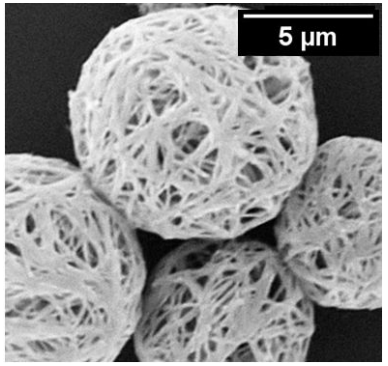
F_r = Rupture force
d = capsule diameter

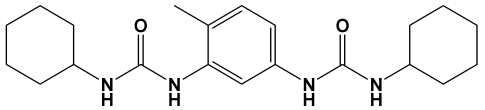
Commercial non-degradable Capsules,
Rupture tension
7 to 111 µN/µm

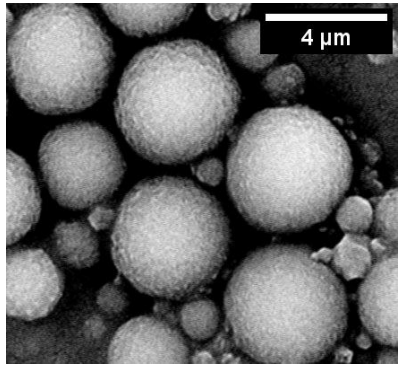
Guinebretiere et. al (2008), WO2008066773A2

Improved Mechanical Strength



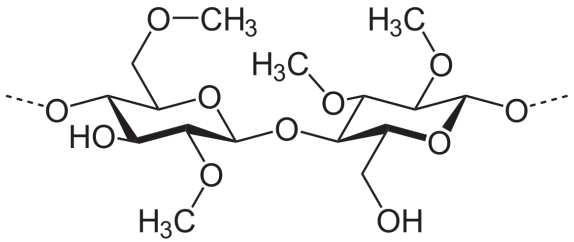


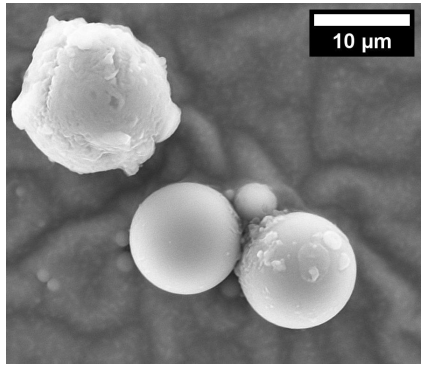




	Rupture tension (µN/µm)
Capsules made using PVA only	**7 ± 1**
Capsules made using PVA and Laponite ®	**23 ± 2**
Capsules made using new molecule	**56 ± 4**

- Methyl Cellulose as an emulsifier

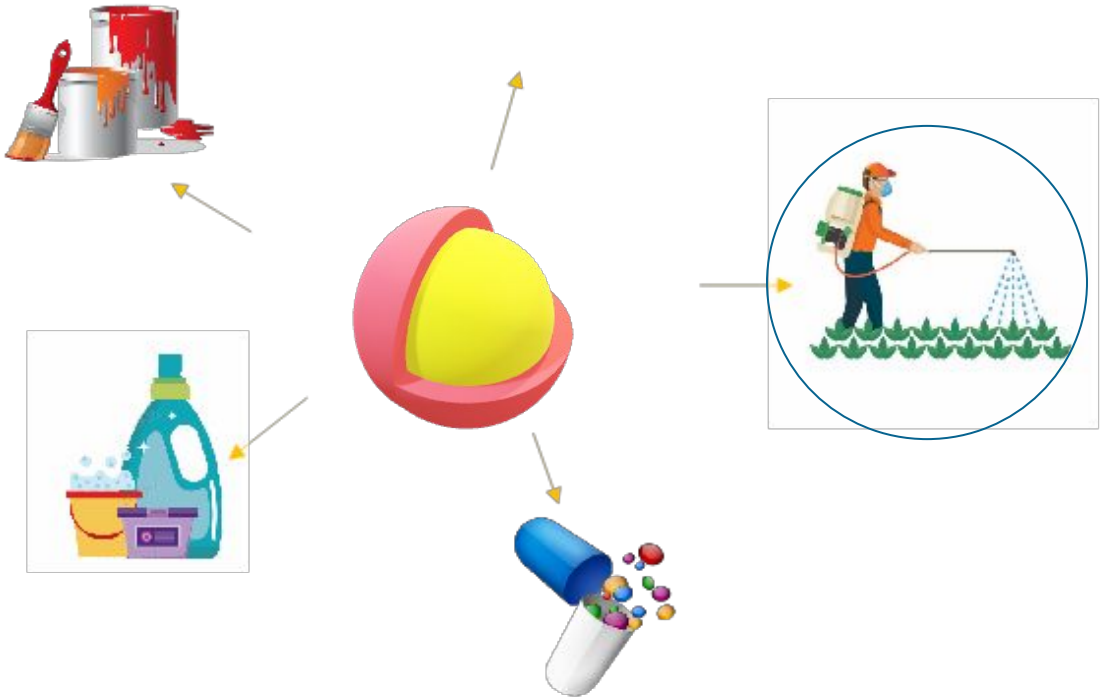




Commercial non-degradable Capsules,
Rupture tension
7 to 111 µN/µm

Guinebretiere et. al (2008), WO2008066773A2

Impact



Global microcapsule market **US$ 11 bn**

- Novel method to synthesize microplastic free microcapsules
- A facile one-pot, *in-situ* process under benign conditions
- Successful encapsulation of commercial herbicide


(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property Organization
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- A plethora of small molecules to test
- Industrial viability: using existing equipment

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