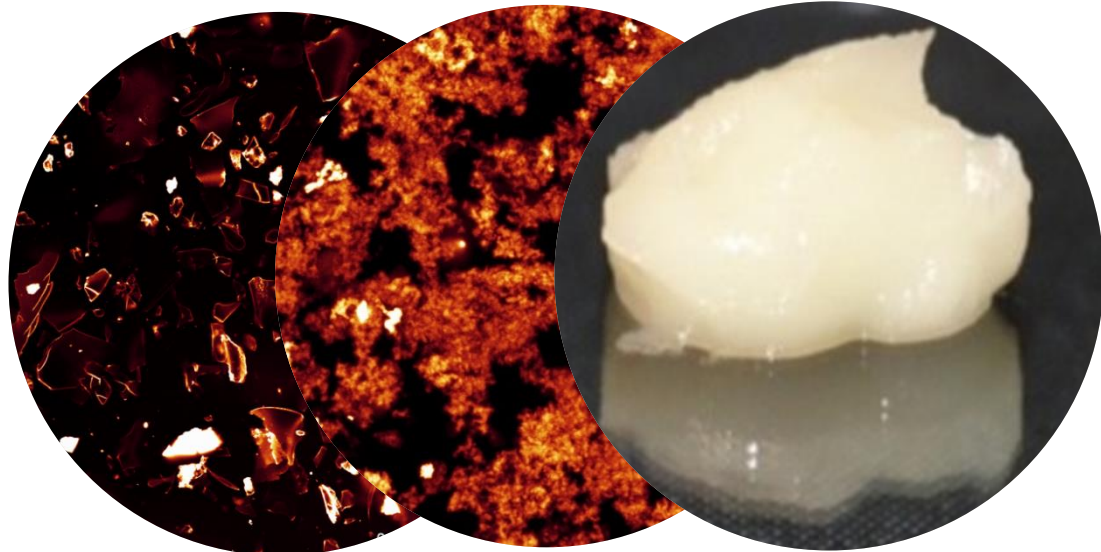
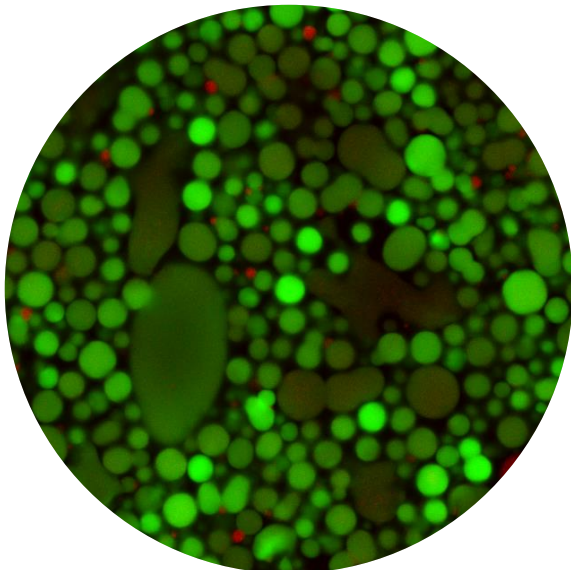


Health and sustainability: Fundamental aspects of oil structuring for different applications

Elke Scholten

Physics and Physical Chemistry of Foods

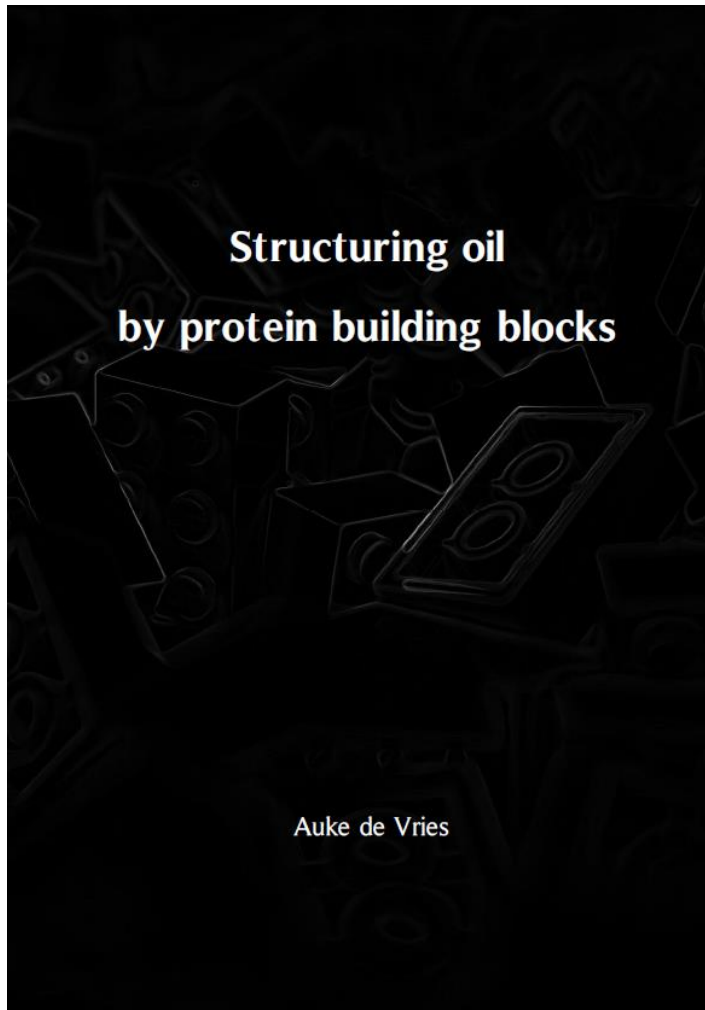
Wageningen University, the Netherlands



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PhD thesis (FS002)



PhD thesis:

Auke de Vries

(graduated March 2017)



- De Vries, A., Hendriks, J., van der Linden, E., Scholten, E. *Langmuir* **2015**, 31, 13850.
- De Vries, A., Wesseling, A., van der Linden, E., Scholten, E. *Journal of Colloid and Interface Science* **2017**, 486, 75.
- de Vries, A., Lopez Gomez, Y., van der Linden, E., Scholten, E. *RSC Advances* **2017**, 7, 11803-11812.
- de Vries, A., Lopez Gomez, Y., Jansen, B., van der Linden, E., Scholten, E. *Applied Materials and Interfaces* **2017**, 9, 10136-10147.



Fat Functionality



Functionality of (solid) fat:

Texture:

- *Layered phase* Hardness
Crispiness
- *Dispersed phase* Hardness
Creaminess

Flavour:

- Hydrophobic flavours

Health:

Saturated fatty acids are assumed to have negative health affects (debated):
WHO → reduction in solid fats: use of unsaturated fats (oil) is encouraged

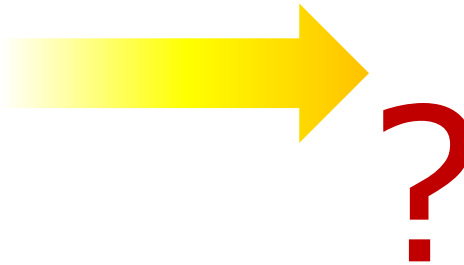
Sustainability

Solid fats: Palm oil, Shea butter, Cocoa butter, Milk fat
Need for alternatives for hard fats

How to make oil act as a solid fat?



Liquid



Solid

Oil composition

Unsaturated fatty acids

→ No detrimental health benefits

Functionality

Provides solid-like characteristics

Controls textural properties such as creaminess, hardness

Still contains fat flavour characteristics

(compared to other fat replacers)

Gelation of oil using structuring agents: **Oleogels**

Resemble fat characteristics:

Solid at small deformation

Yield at larger deformation (break-down)

Melting behavior



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Oil Gelation

Known Categories

Particle Networks (fats..)

Low molecular weight surfactants

Self assembly behavior:

- *Lecithin*
- *sterols*
- *waxes*

Poly

Netv
Enta

- *Ethyl cellulose*
- *chitin*



Oleogel / organogel

Continuous phase of oil
Network of fillers

- **Not healthy**
- **Not efficient**
- **Too expensive**
- **No legal status**
- **Etc..**

Requirements for foods:

- *Natural origin*
- *Cheap*
- *Widely-available*

New Category

Protein oleogels

Proteins hydrogels

Yogurt
Cheese
Eggs
Desserts
Tofu

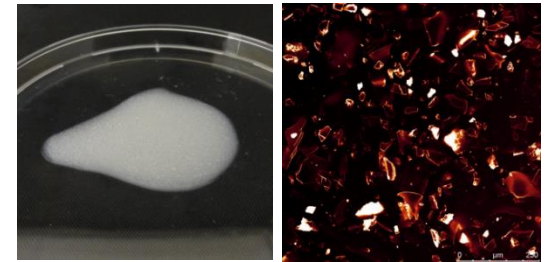
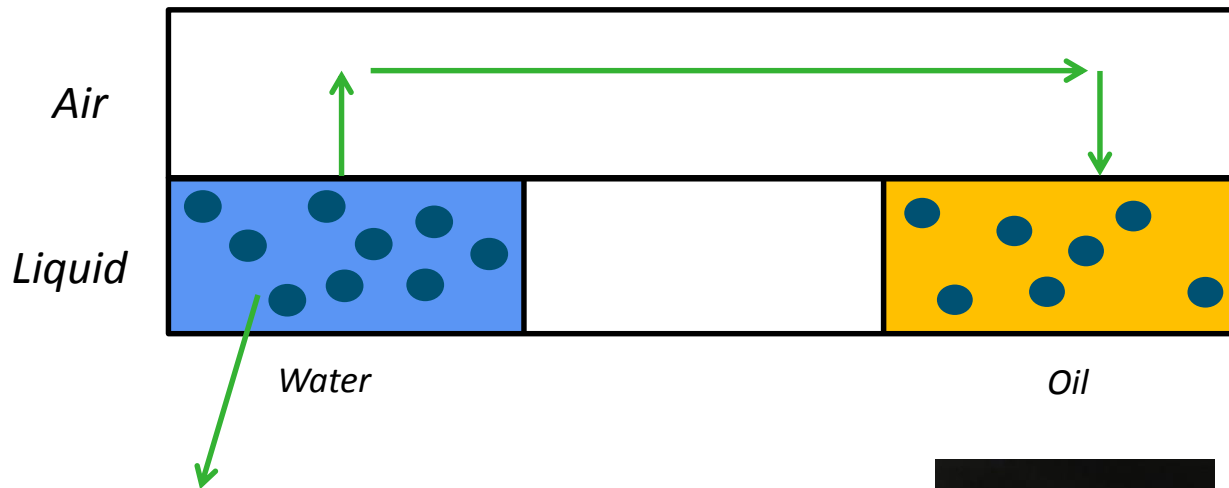


Protein based oleogels

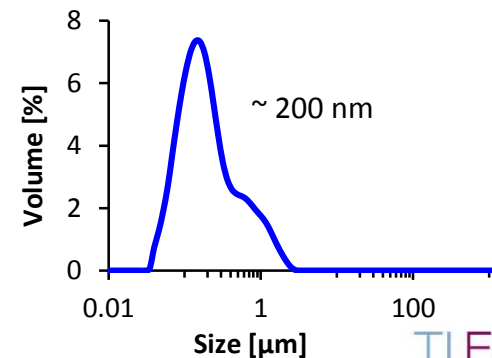
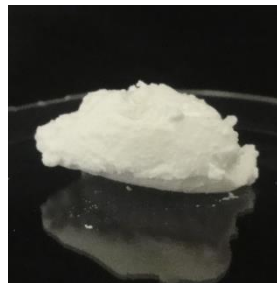
Proteins well-known for ability to create hydrogels in aqueous environments

How to get them in the oil phase ?

Sedimentation of proteins



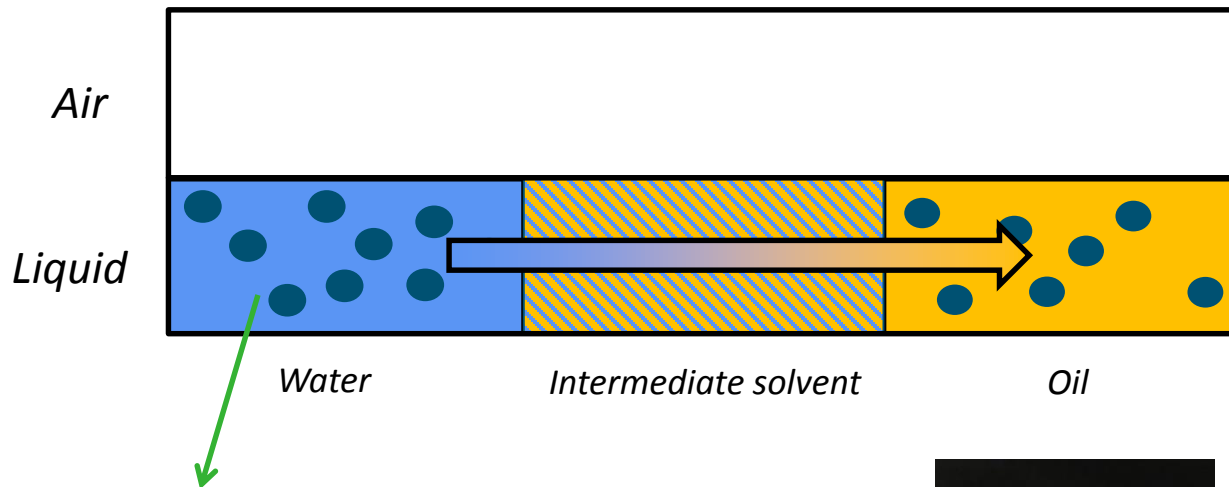
Modification : heat treatment (denaturation)
create initial **building block**
increased hydrophobicity



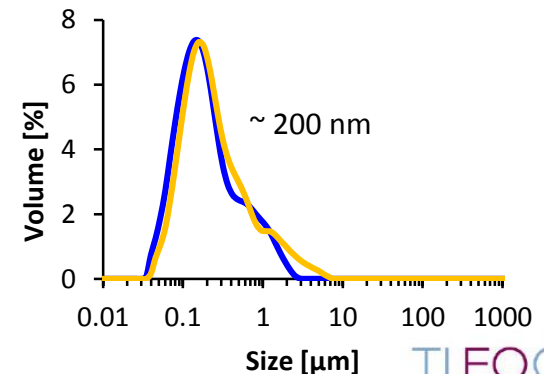
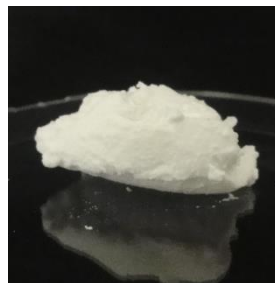
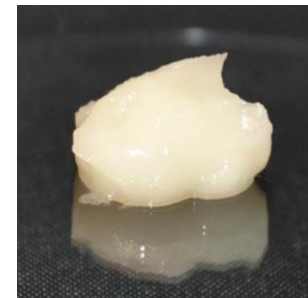
Protein based oleogels

Proteins well-known for ability to create hydrogels in aqueous environments

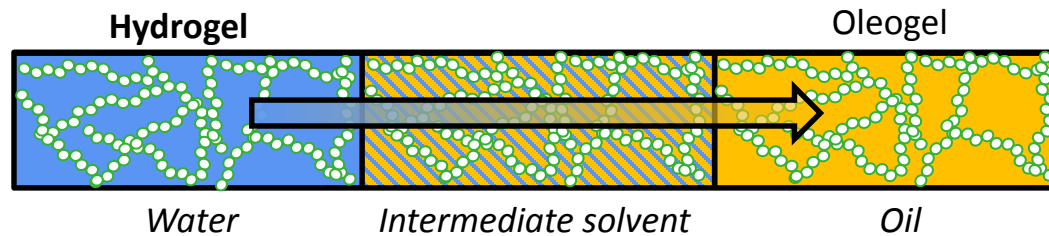
How to get them in the oil phase ?



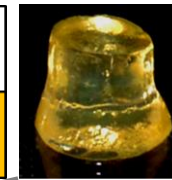
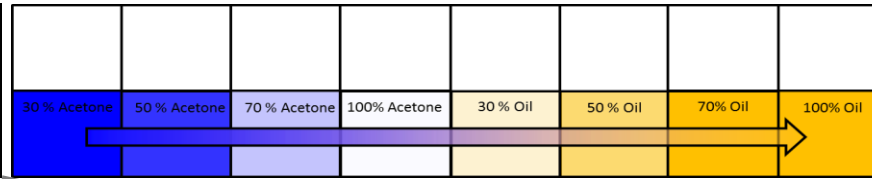
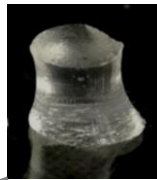
Modification : heat treatment (denaturation)
create initial **building block**
increased hydrophobicity



Protein oleogels – from hydrogel (mm – cm)



Cm scale



~ 5% Protein
~ 91% Oil
< 1% Water

Microstructure design

*Fine -
stranded
Transparent*



*Coarse
Opaque*

← →
pH, salt, type of protein

oleogels

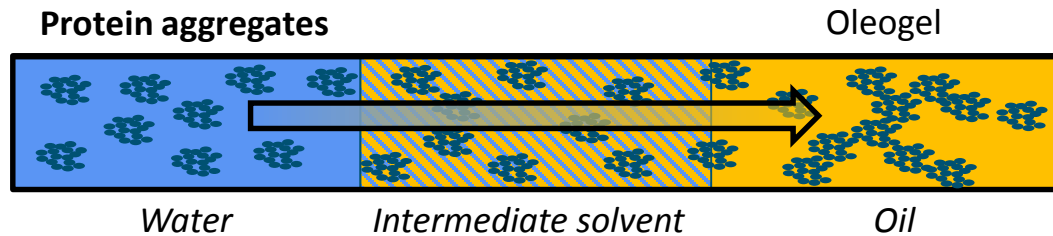
*Fine -
stranded
Transparent*



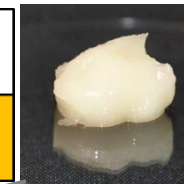
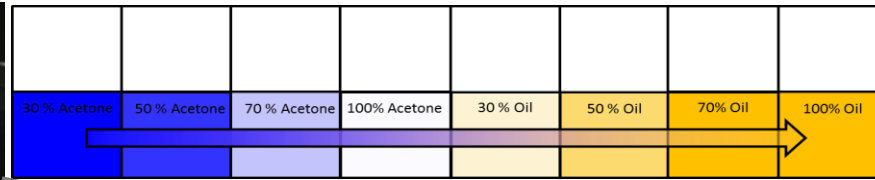
*Coarse
Opaque*

Protein content
Oil content

Protein oleogels – from protein aggregates (nm)



nm scale



~ 5% Protein
~ 91% Oil
< 1% Water

Microstructure design

pH
Salt

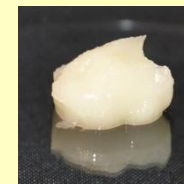
Protein type
(plant-based)



Size
Density
Hydrophobicity

oleogels

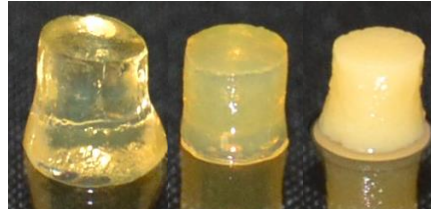
Concentration



Protein content
Oil content
Network

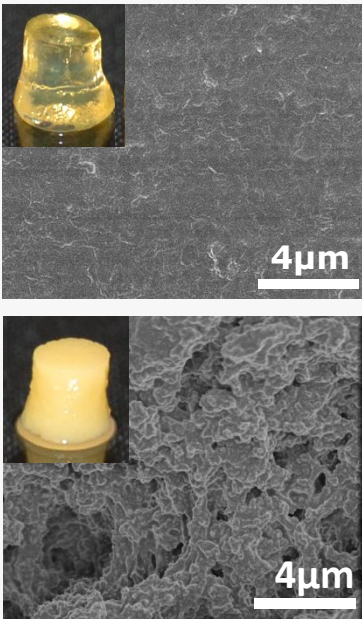
Characteristics (from hydrogels)

5 mm



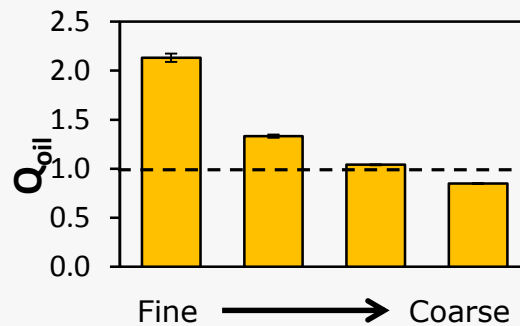
91% - 80% Oil
5% - 16% Protein

Microstructure



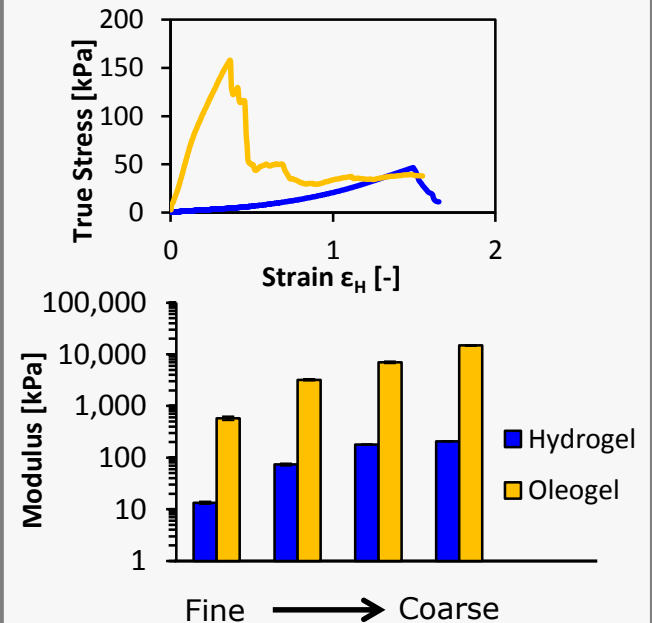
Oil holding

Self standing gels
No oil-leakage



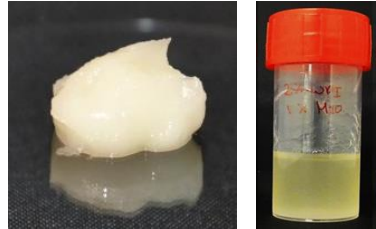
Oil binding depends on the microstructure

Mechanical properties



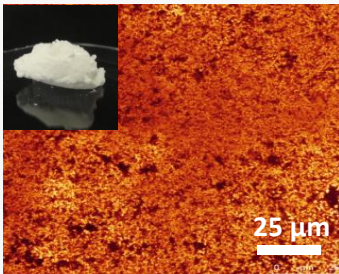
Characteristics (*from aggregates*)

5 mm

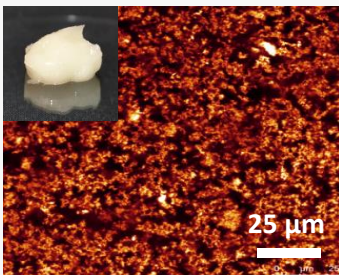


Varying protein content

Microstructure

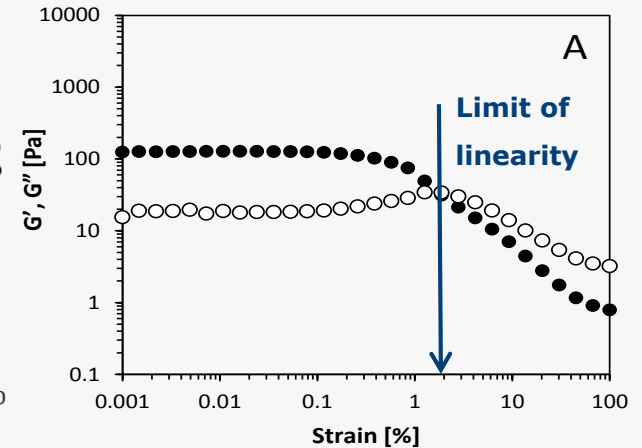
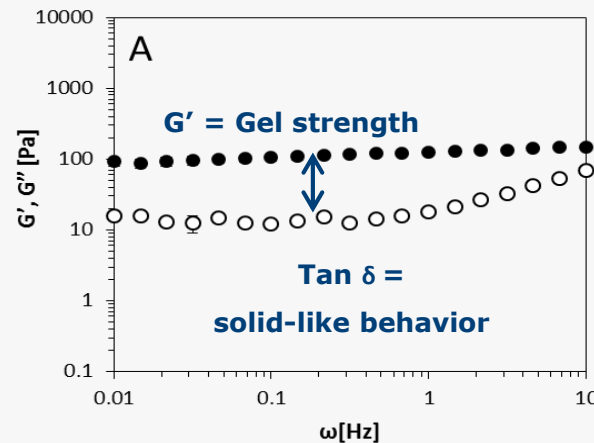


In Water



In Oil

Mechanical properties



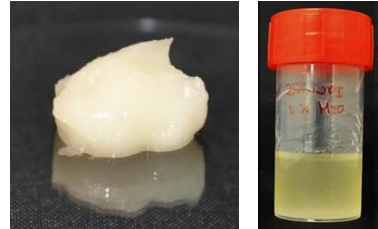
No oil leakage

Show solid-like behavior

Show yield behavior → spreadable

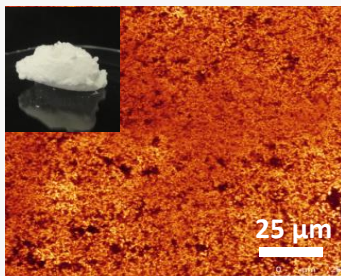
Characteristics *(from aggregates)*

5 mm

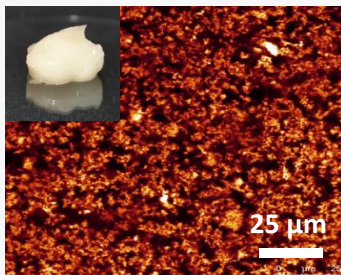


Varying protein content

Microstructure

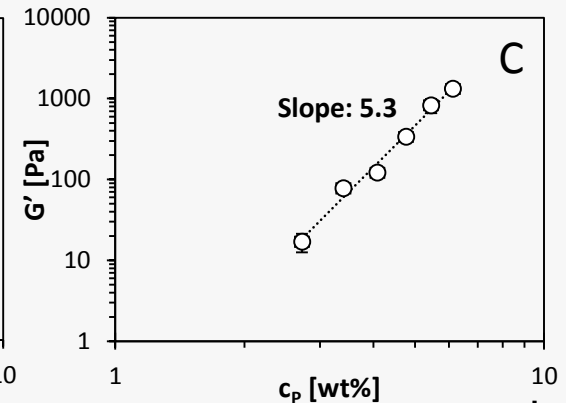
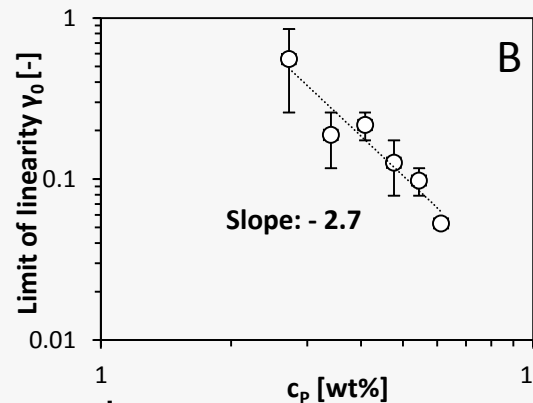


In Water



In Oil

Network formation



Fractal gel model: "Strong-link network"

**Protein network same
as in water**

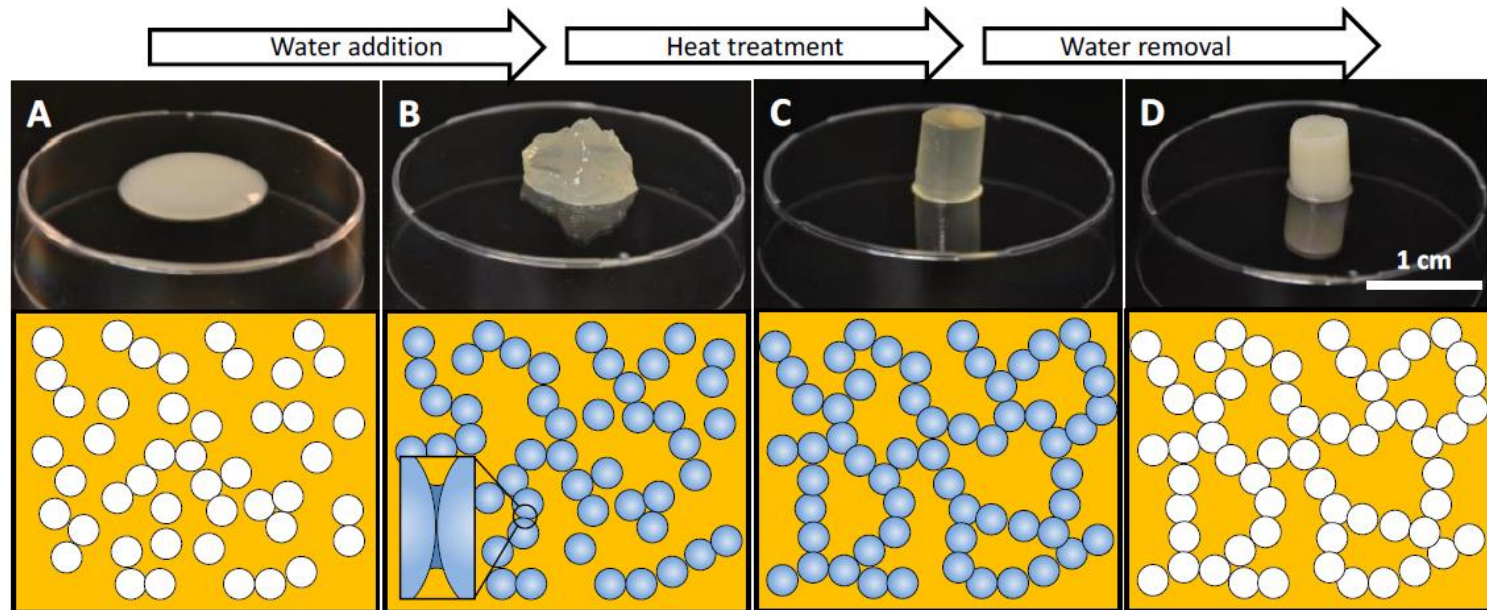
$$G' \sim \varphi^{(d+x)/(d-d_f)} \longrightarrow d_f \sim 2.2$$

$$\gamma_0 \sim \varphi^{-(1+x)/(d-d_f)}$$

Shih et al. *Phys Rev A*, 1990

Network formation - *Rheological properties*

Control network → gel strength, yield stress, plastic deformation



Water addition:
hydrogen bonding
capillary interactions

Heat treatment
hydrogen bonding
capillary interaction

Water removal
Hydrogen bonding
Van der Waals interactions

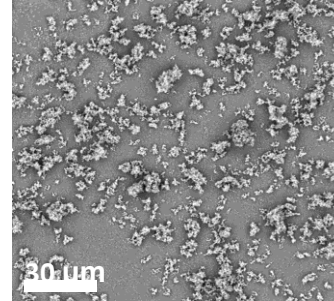
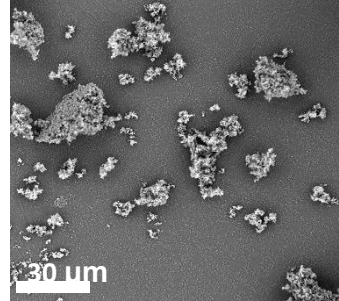
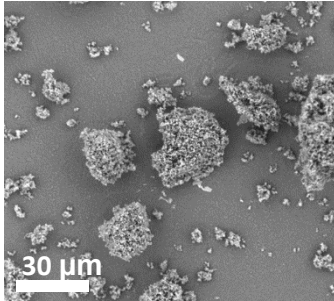
**Rearrangements in the
network**



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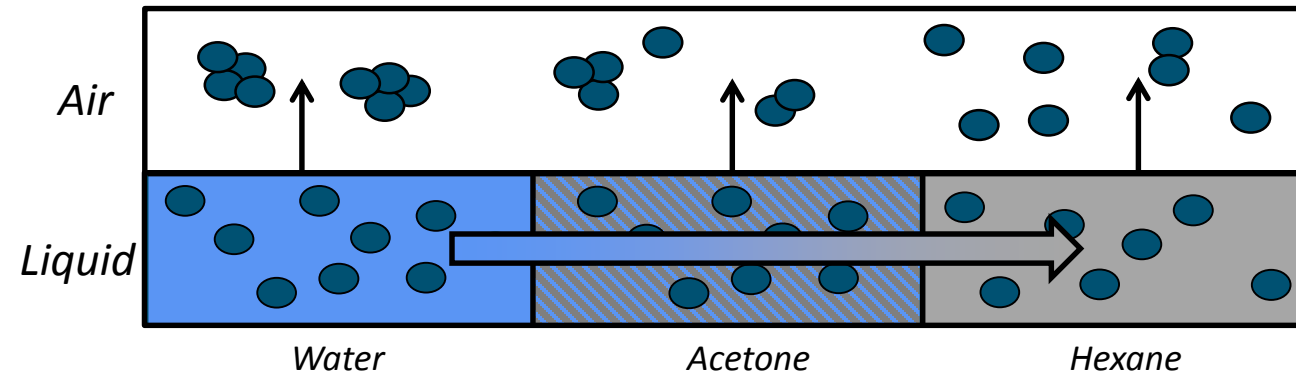
Upscaling ??- *Drying from different solvents*



Solvents prevent agglomeration

(hydrophobicity, interfacial tension)

→ Better dispersibility in oil



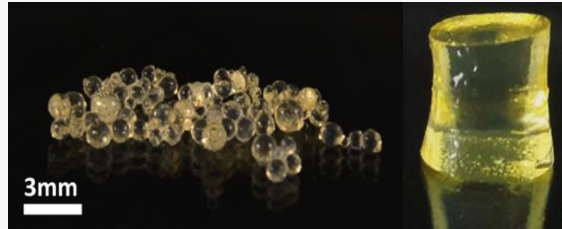
How to accomplish ??

- Drying from organic solvents
- Optimization in freeze drying process
- CO₂ drying ?

→ Dry protein aggregates powder

Applications

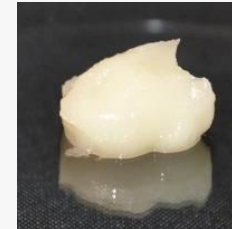
Protein hydrogel



Fracture properties
Gel strength

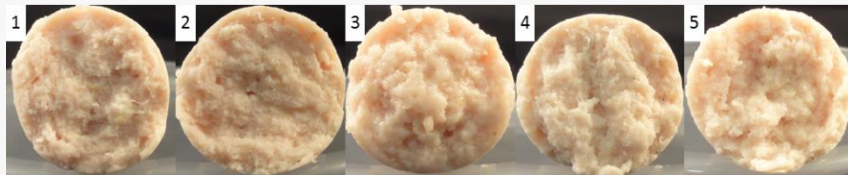


Protein aggregates



Spreadable properties
Yield behavior

Sausages



Fat

Fat

Oil

Aggregates

Pieces

Cookies



Margarine

Aggregates

Aggregates

Oil

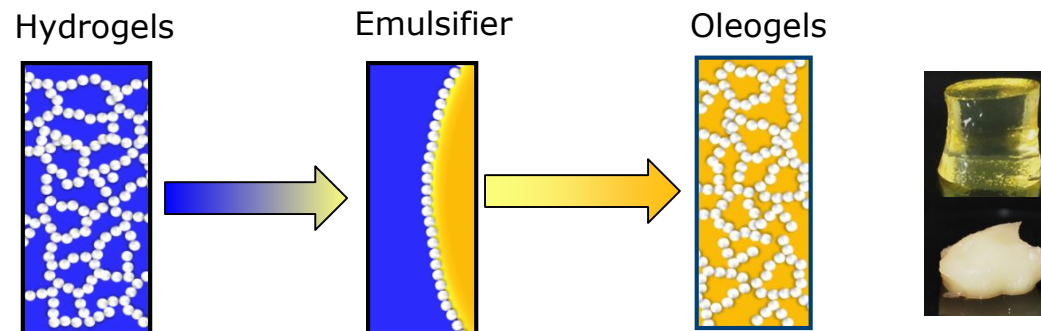
Protein oleogels have potential as a fat replacer

Conclusion



Gelation of oil with proteins:

- ✓ Healthy composition with solid character
- ✓ Tunable oleogel characteristics/properties (*applications*)
- ✓ Interactions can be controlled by:
 - Aggregate properties (size, hydrophobicity, source, density)
 - Interactions: water addition and heat treatment
- ✓ Can be used as alternative for solid fats (palm oil, shear butter, milk fat)



New type of protein functionality: ***Oil gelation***