Design of a multi-stage membrane filtration system for concentration and separation of colloids: example of skim milk microfiltration

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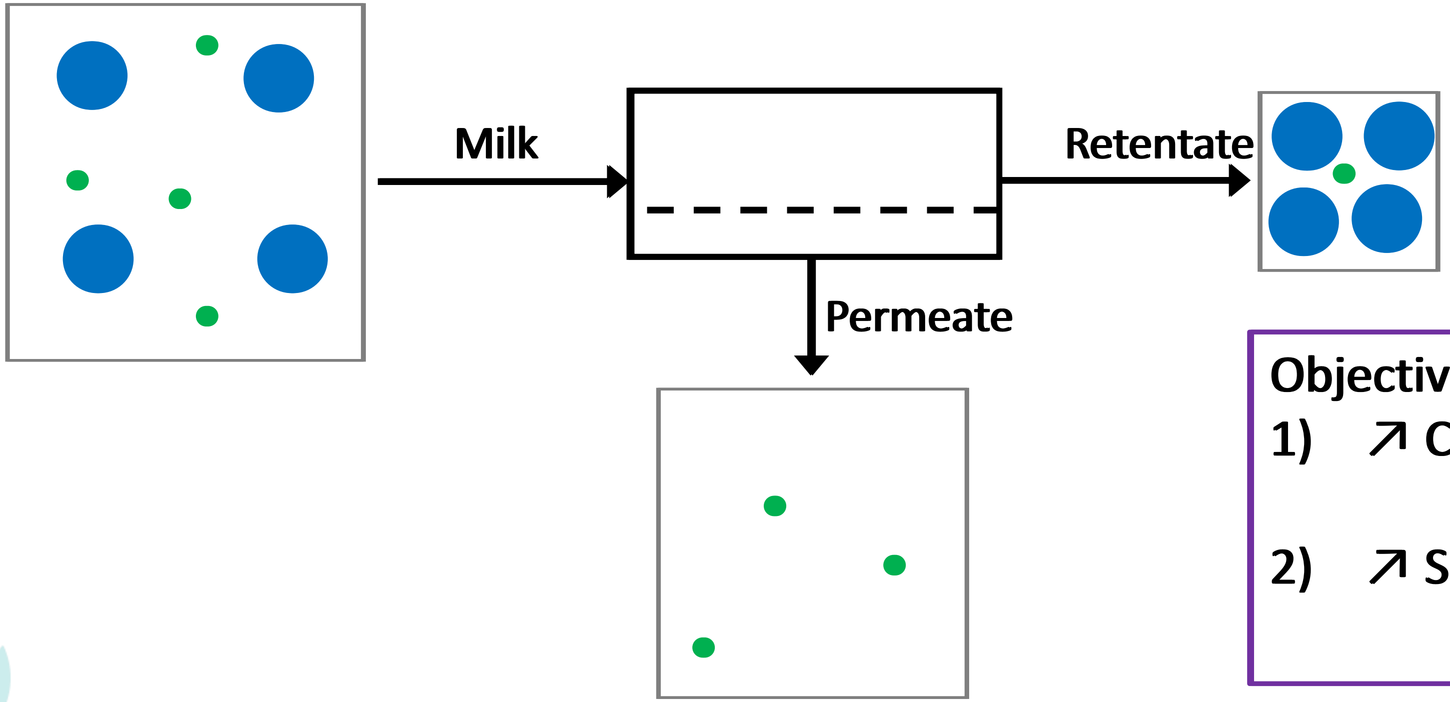
Concentration-separation, example of skim milk microfiltration (0.1 µm)

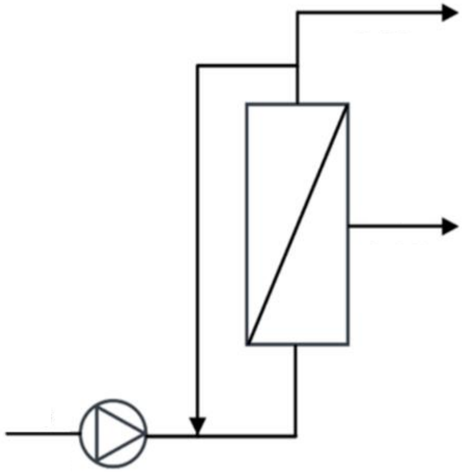
- Microfiltration and ultrafiltration are widely applied in the food sector for concentration or separation of micro or nano-colloids
- Example: MF 0.1 µm of skim milk : Concentration of casein micelles and separation of serum proteins
  Skim milk = Casein Micelles & Serum Proteins
  CM ≈ 150 nm   SP ≈ 5 nm

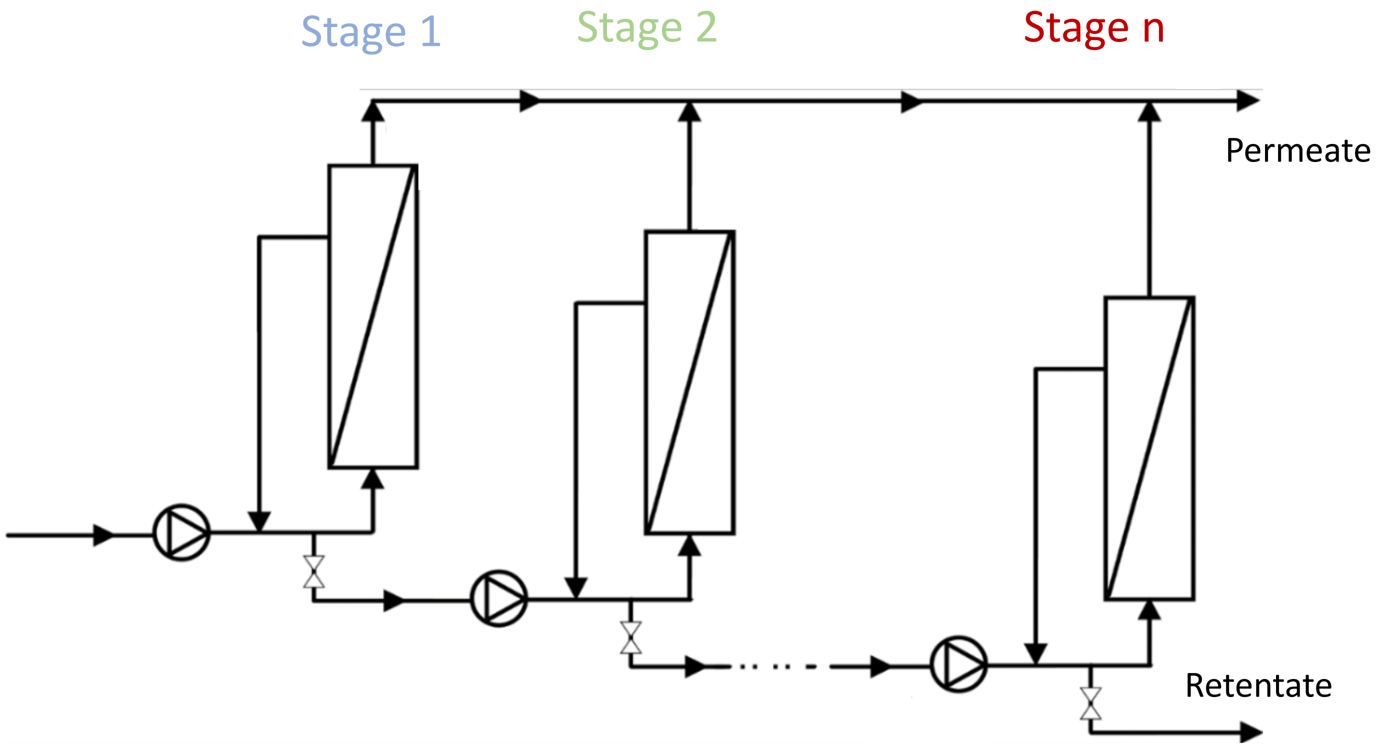


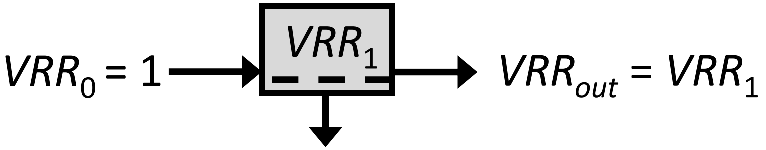
Objectives:
1) ↗ Concentration of CM (high VRR)
   → Cheese making
2) ↗ Separation of SP from CM
   → Ingredients

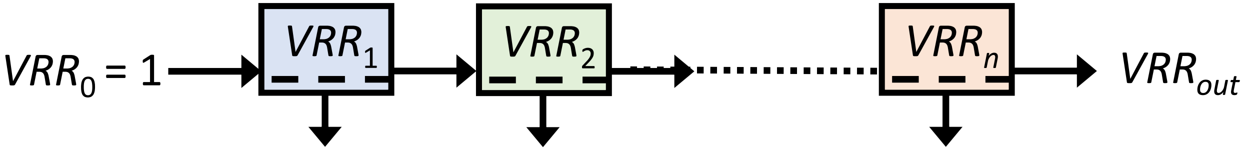
Industrial design of MF and UF in food sector

- Continuously operating system
- N successive stages









<u>For skim milk MF (ceramic membrane; $VRR_{out} \approx 3.5$)</u>

3 stages

Same operating conditions for each stage

Same membrane surface for each stage

Advantages:

- Reduction of membrane surface
- Reduction of residence time of product:
  ↘ denaturation; ↘ development of bacteria

Design of multi-stage membrane filtration systems / Objective

➢ The design rules are not fully established and shared

- The design is made by equipment manufacturers and is mainly based on confidential data and know-how (equipment manufacturers want to preserve their know-how).

➢ Improvements in the design are still possible

- The design is based on simplifying assumptions (and rarely take into account the relation between the process efficiency and the operating parameters)

➢ There is a need for optimization of multi-stage membrane filtration design

  - To compare various configurations
  - To integrate environmental criteria

Propose a general methodology to design the multi-separation-concentration system

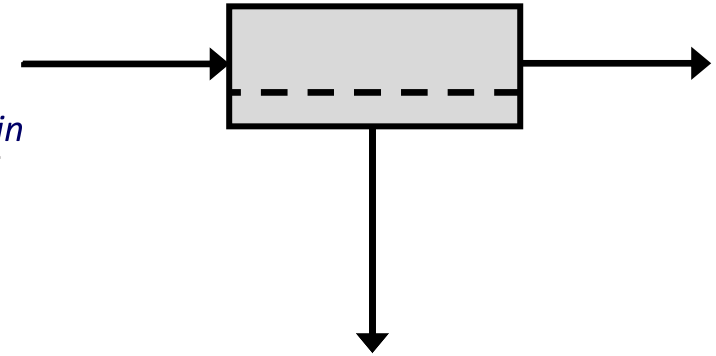
Objective 1: Minimizing the membrane surface for a system operating at desired final retentate (CM) concentration (VRR_{out})

Objective 2: Maximising the solute (SP) recovery yield in the permeate

Objective 1 : Minimizing the membrane surface area, S : Mass balance

Input flow rate Q_{in}
Input concentration $[CM]_{in}$

$VRR_{in} = 1$



Output flow rate Q_{out}
Output concentration $[CM]_{out}$

Volume reduction ratio VRR
$VRR_{out} \equiv Q_{in}/Q_{out} = [CM]_{out}/[CM]_{in}$

Permeate concentration $[CM]_p = 0$ (hyp : Retention of CM = 100%)
Permeate flow rate Q_p
Permeate flux J_p

Membrane surface area S
$\mathbf{S = Q_p/J_p}$

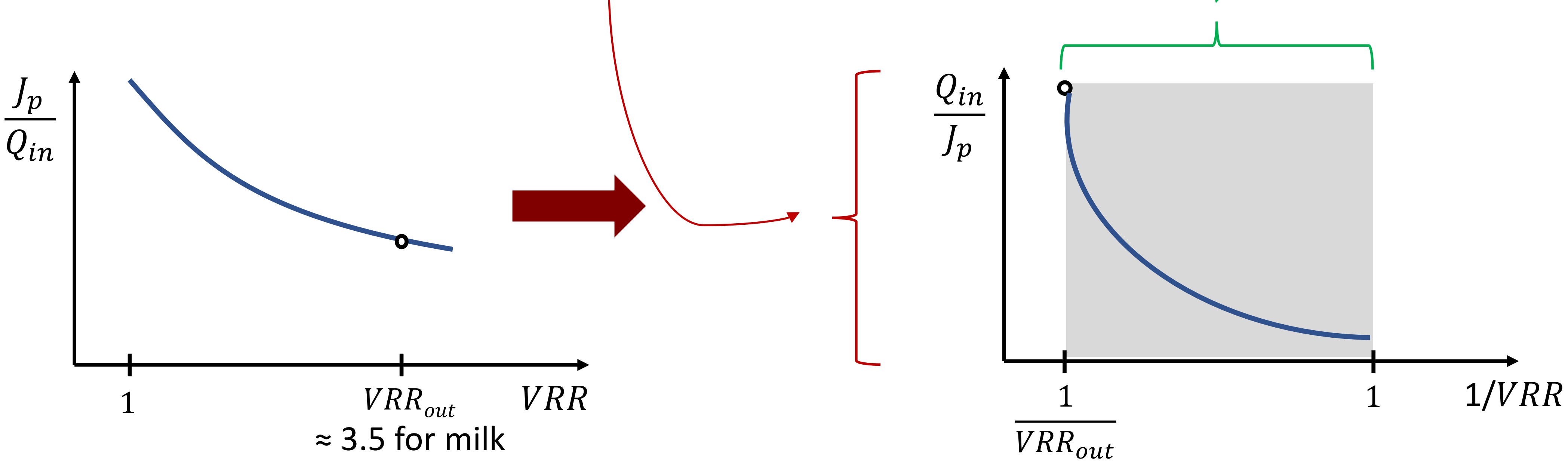
$$S = \frac{Q_{in}}{J_p}\left(\frac{1}{VRR_{in}} - \frac{1}{VRR_{out}}\right)$$

(Jeantet, Brulé, et Delaplace 2011)

Objective 1 : Required membrane surface area, S

$$S = \frac{Q_{in}}{J_p}\left(\frac{1}{VRR_{in}} - \frac{1}{VRR_{out}}\right)$$

N=1



Higher VRR_{out} → lower J_p → higher S

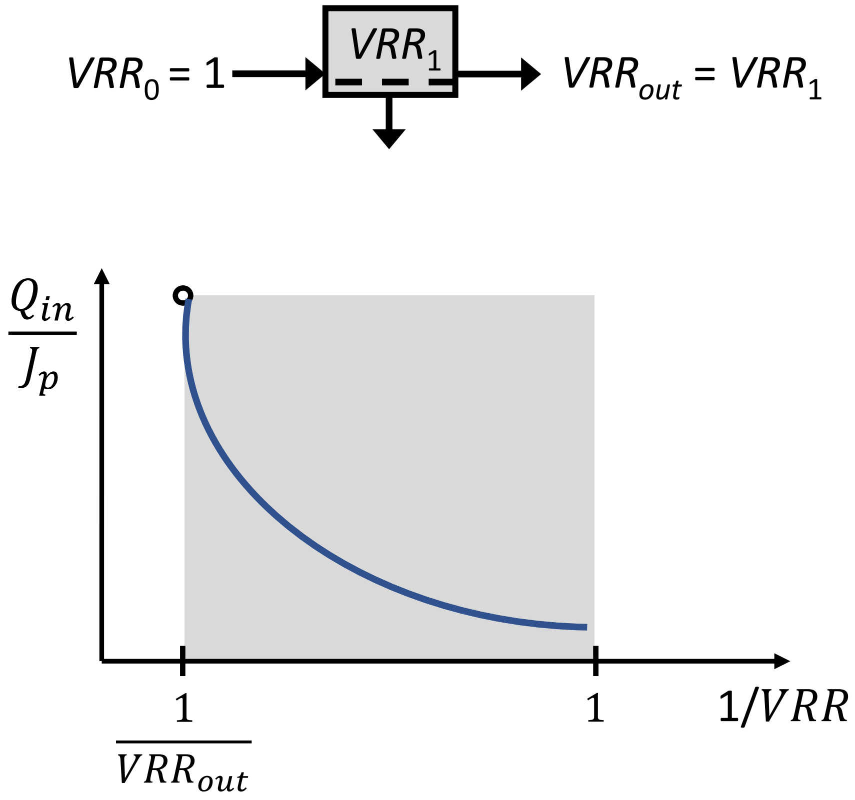
S = $!

S = surface of the shaded rectangle

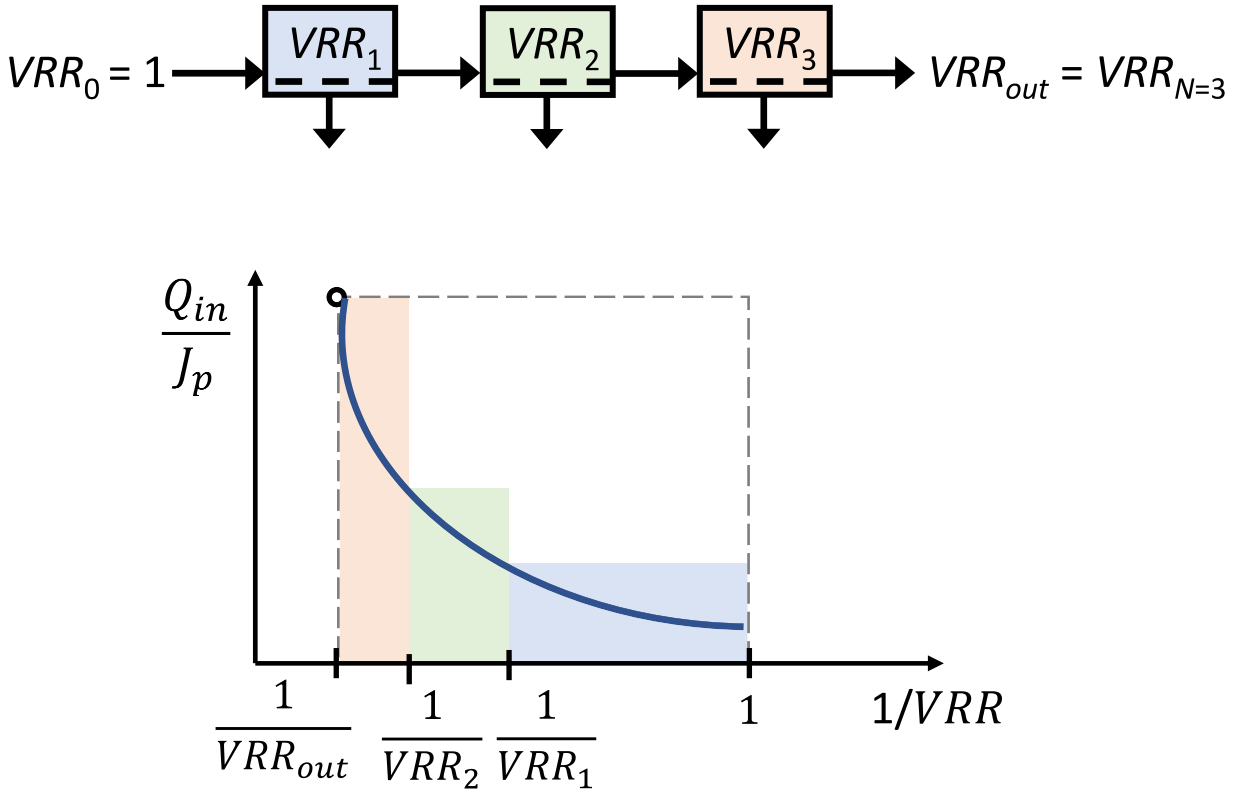
(Jeantet, Brulé, et Delaplace 2011)

Objective 1: Interest of a multistage filtration system

N=1

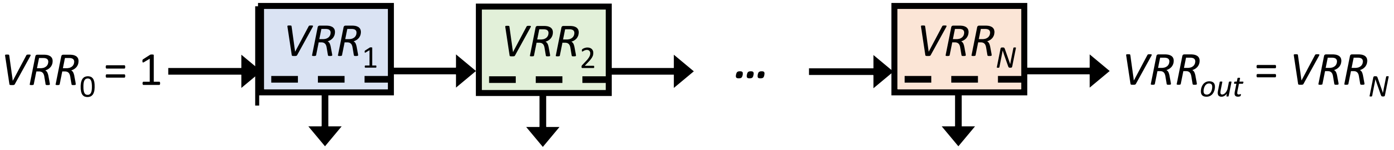


N=3



J_p decreases with VRR → multistage system requires less total S (S_Σ) than 1-stage system
More stages (N) → lower required total S_Σ
Limitation by the cost of equipment

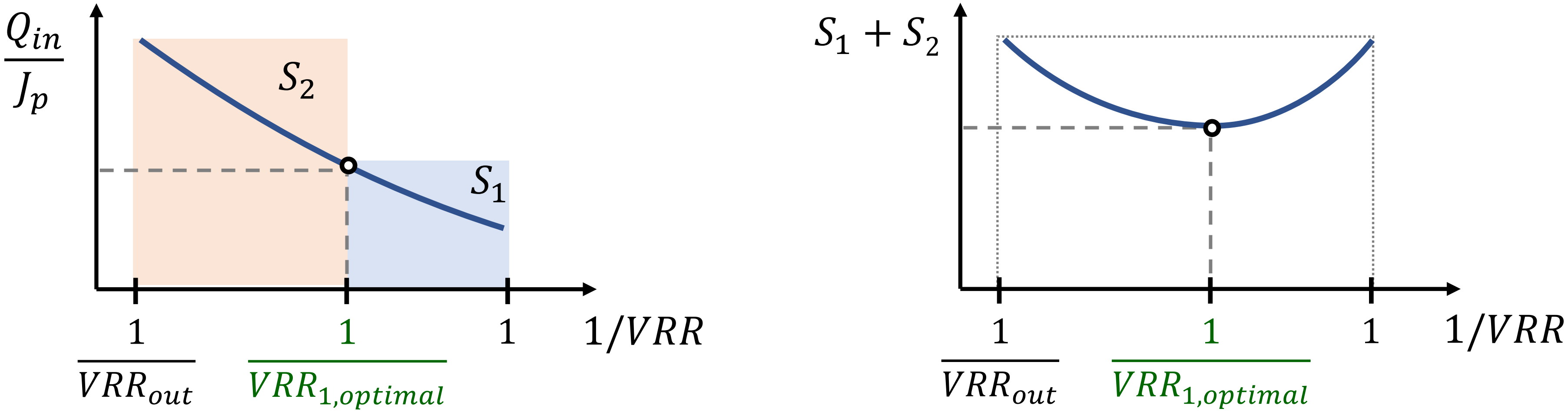
Objective 1: Minimization of total membrane surface area, S_Σ



$$S_{\Sigma,min} = \min_{VRR_i} \sum_{i=1}^{N-1} \left(\frac{1}{J_{p,i}} \left(\frac{1}{VRR_{i-1}} - \frac{1}{VRR_i} \right) \right)$$

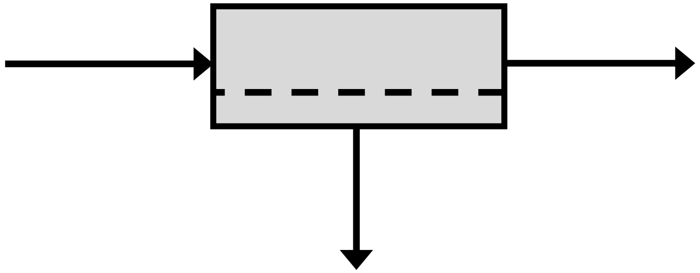
- Optimal values of VRR_i ($i = 1..N-1$) for given Q_{in}, $[CM]_{0\ \&\ N}$, required VRR_{out}, and given $J_p = f(VRR)$;
- Values of S_i ($i = 1..N$)

Illustration for two-stage filtration: determination of optimal VRR_1



Objective 2: Maximizing the recovery yield of serum proteins in permeate, *Y*

Input flow rate Q_{in}
Concentration of SP $[SP]_{in}$



Retentate flow rate Q_r
Concentration of SP $[SP]_r$

Permeate flow rate Q_p
Concentration of SP $[SP]_p$

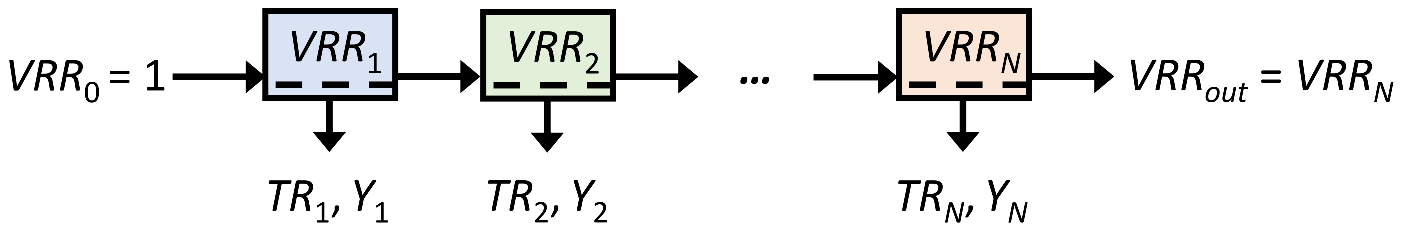
Transmission of SP *TR*

$\mathbf{TR = [SP]_p / [SP]_r}$

Recovery yield of SP in permeate *Y*

$\mathbf{Y = [SP]_p Q_p / [SP]_{in} Q_{in}}$

Objective 2: Yield of SP vs. configuration of a multi-stage filtration system : Mass balance



Total yield of SP in permeates, Y_Σ

$$Y_\Sigma = \sum_{i=1}^{N} Y_i \quad \text{where} \quad Y_i = \frac{1}{VRR_{i-1}} \prod_{j=0}^{i-1} b_j - \frac{1}{VRR_i} \prod_{j=0}^{i} b_j \;;$$

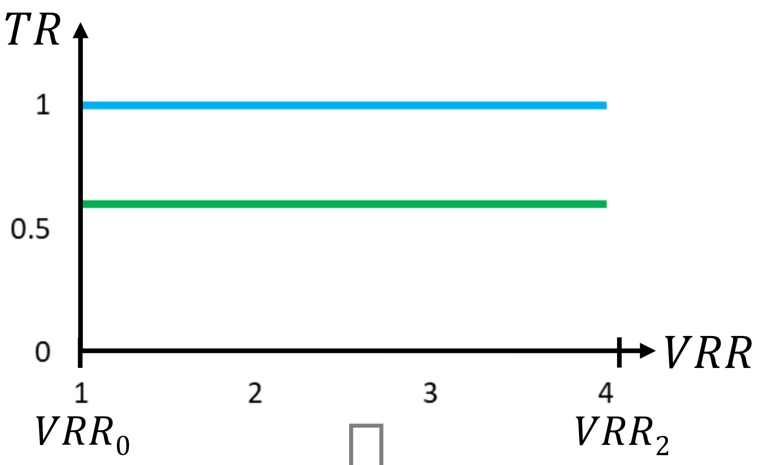
$$\text{with} \quad b_j = \frac{VRR_j}{TR_j\left(VRR_j - VRR_{j-1}\right) + VRR_{j-1}}$$

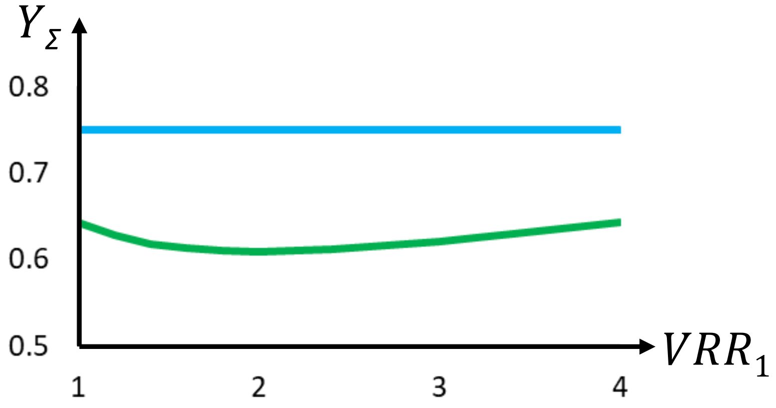
NB: (1) Y_i depends on all VRR_j and all TR_j ($j = 1..i$)

(2) $TR = f(VRR)$

Objective 2 : Total yield Y_Σ vs. TR = f(VRR): Possible scenarios (ex two-stage system)

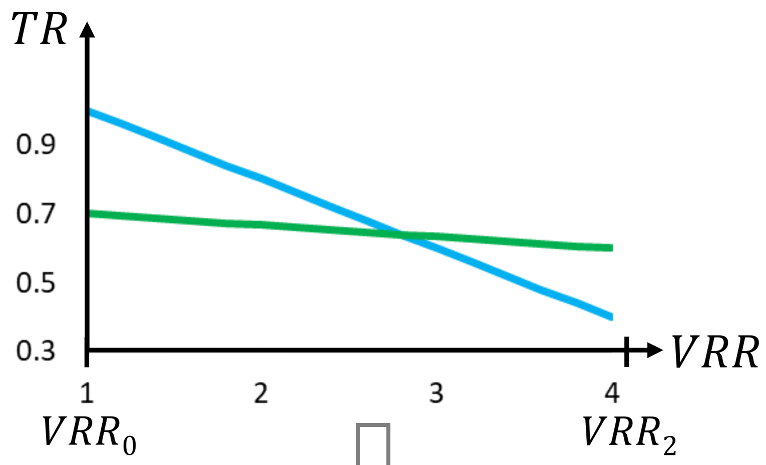
1) *TR* is constant

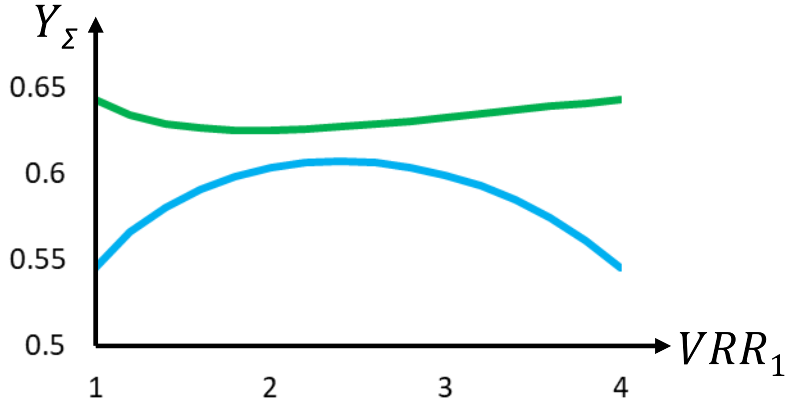




Y_Σ *vs.* VRR_1 can pass through a minimum

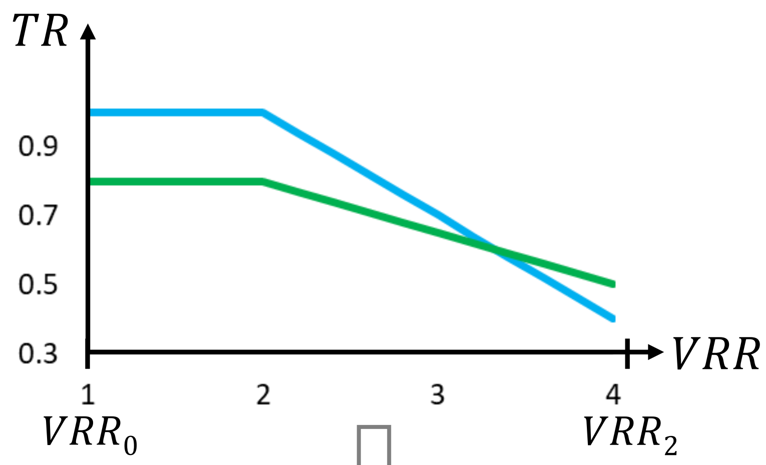
2) *TR* drops linearly with *VRR*

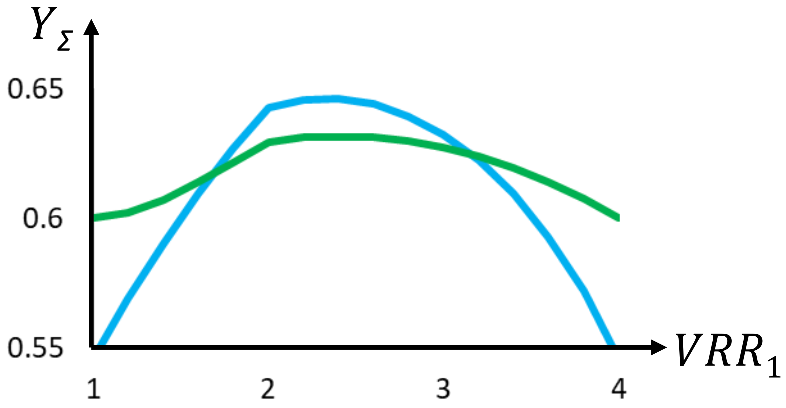




Possible maximum/minimum; higher local *TR* ≠ higher Y_Σ

3) *TR* drops at high *VRR*

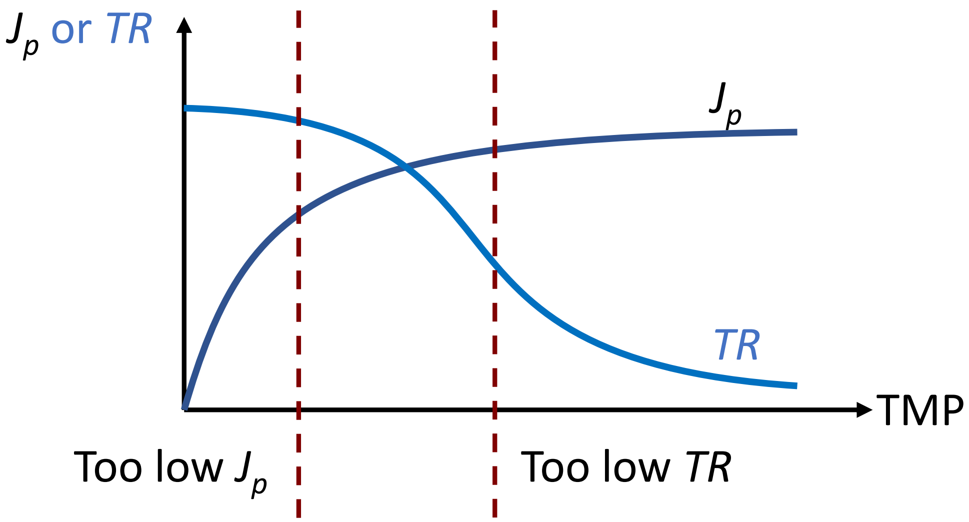




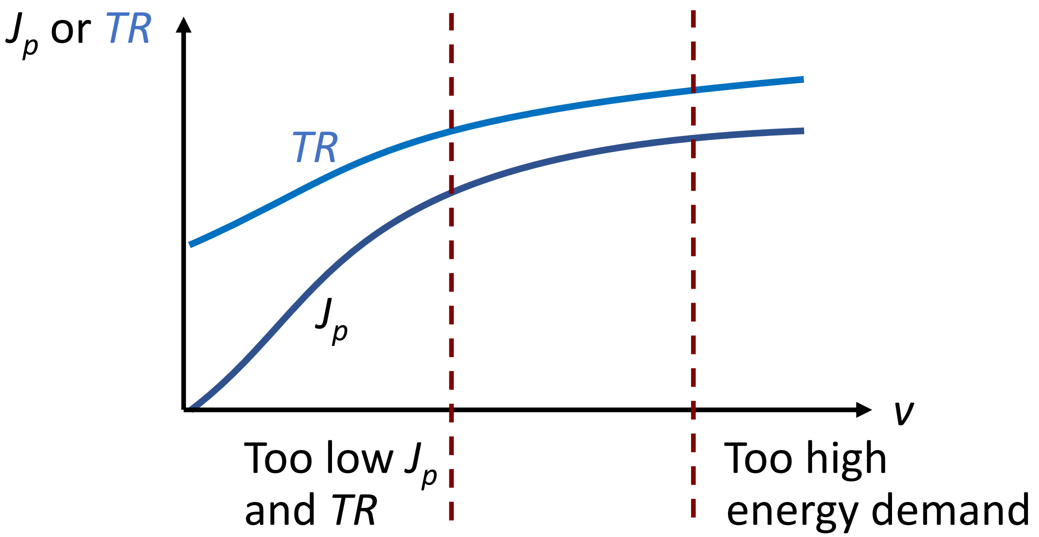
Observed optimal VRR_1 is above the break point

What else can affect Jp & TR = f(VRR) and influence the system design ?

1) Transmembrane pressure TMP



2) Retentate crossflow velocity v



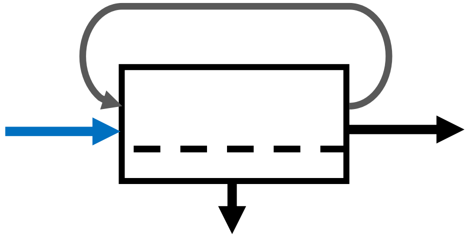
3) Concentration of SP in all retentates $[SP]_{r,i}$

- $[SP]_{r,i}$ at VRR_i depends on all VRR_j, $j = 1..i$
- Influence of $[SP]_r$ on J_p and TR is practically unknown

- $J_p(VRR)$ and $TR(VRR)$ at different TMP and v must be measured for given membrane module
- Experimental study is needed to elucidate the influence of $[SP]_r$ on J_p and TR

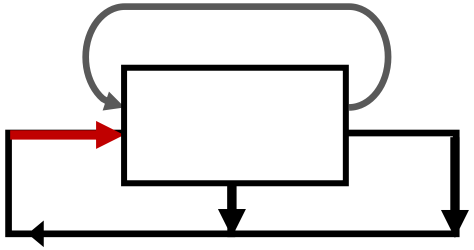
Batch filtration for scale-up of continuous filtration?

Continuous

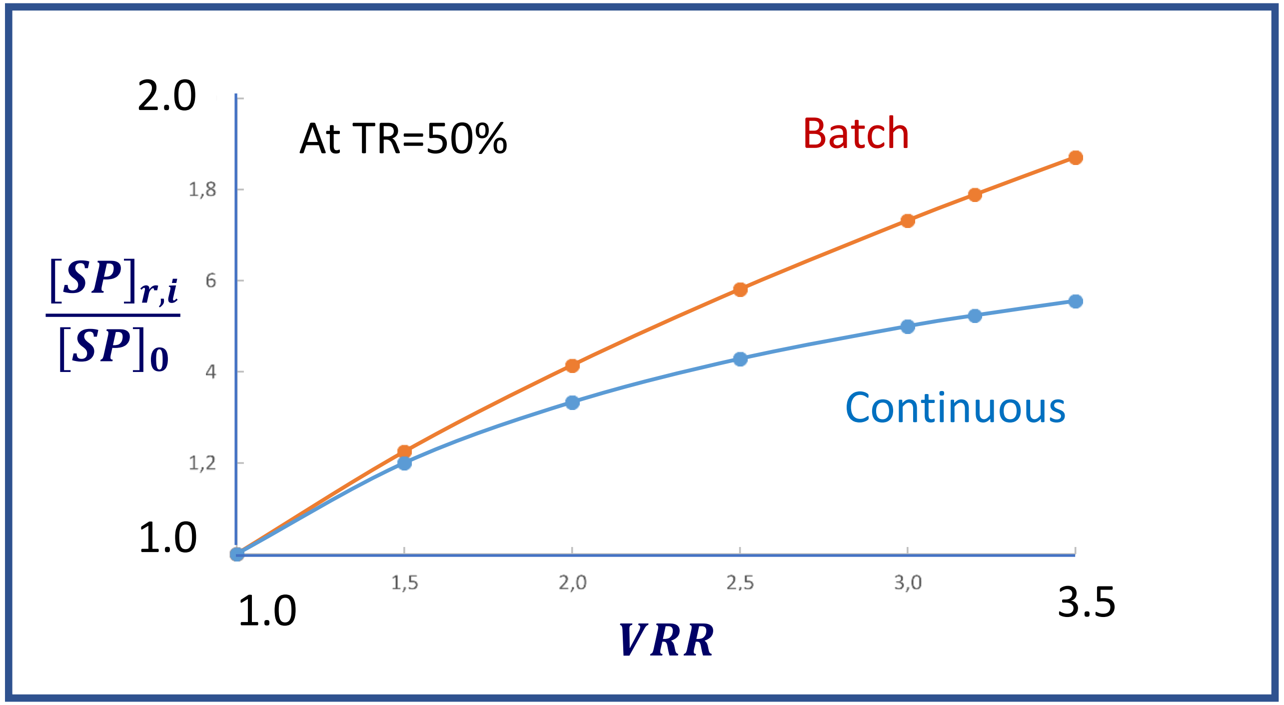


$$\frac{[SP]_{r,i}}{[SP]_0} = \prod_{j=0}^{i} \frac{VRR_j}{TR\left(VRR_j - VRR_{j-1}\right) + VRR_{j-1}}$$

Batch



$$\frac{[SP]_{r,i}}{[SP]_0} = VRR_i^{1-TR}$$



Continuous mode *vs.* batch mode have different $[SP]_r$ at same VRR

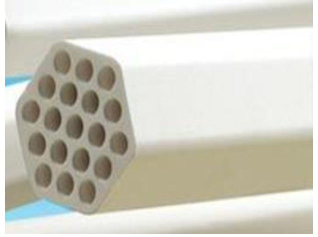
This can result in different dependencies of J_p and TR on VRR

Another reason to verify, whether J_p and TR depend on $[SP]_r$

INRAE Dairy platform, STLO, Rennes

- **Microfiltration pilot** TetraLaval MFS7 equipped with a uniform transmembrane pressure (UTP) system
- **Membrane module** with 7 ceramic membranes Pall 7P1940 GL

membrane length	1.02 m
number of channels	19
channel diameter	4 mm
membrane surface area	1.68 m^2

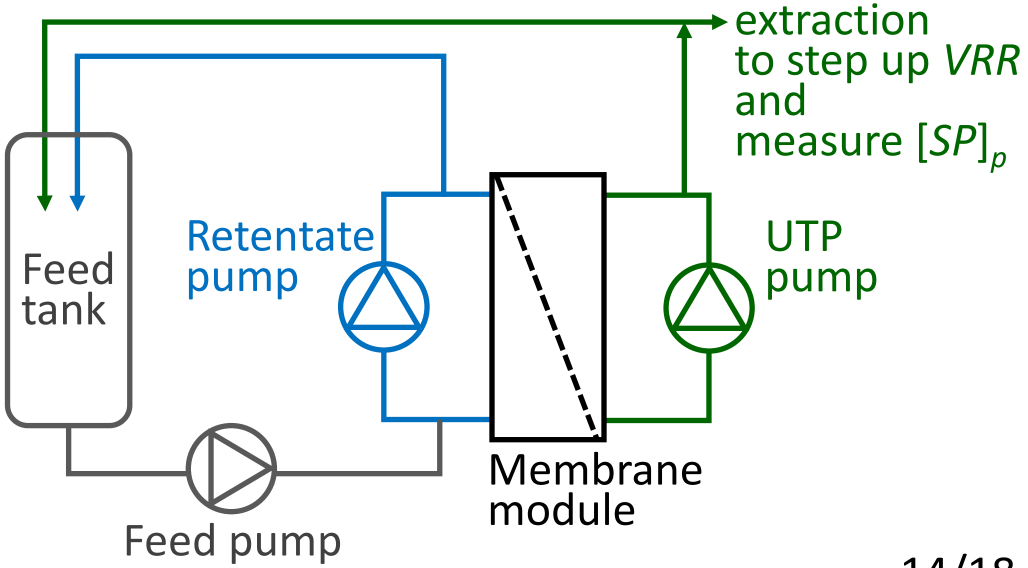


- **Milk** (skimmed, pasteurized) provided by Coralis, Cesson Sévigné (F-35)
- **Milk enriched** with whey proteins concentrate (WPC):

$$[SP]_{\text{enriched milk}} = 2 \times [SP]_{\text{milk}}$$

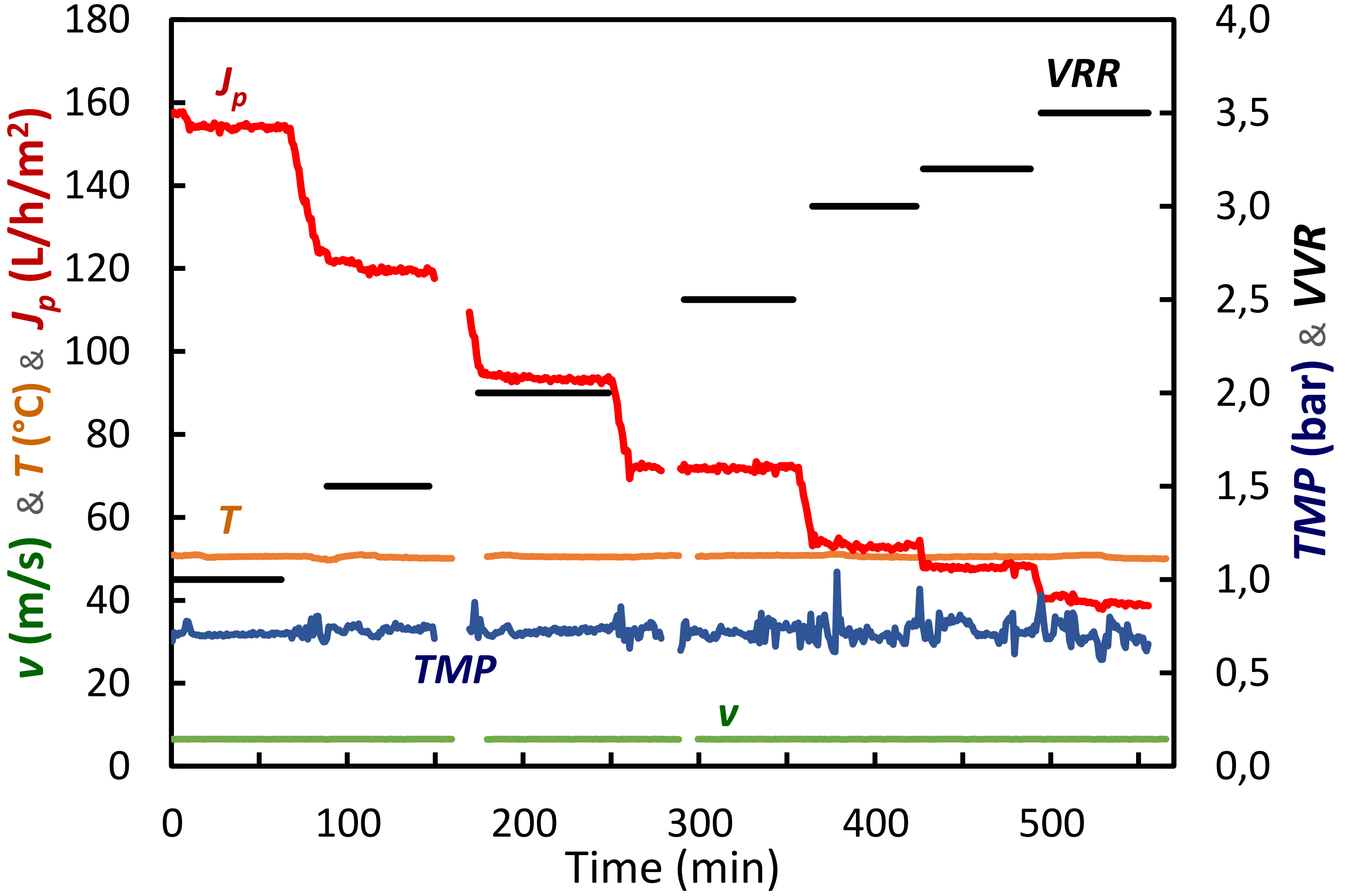
to obtain J_p and *TR vs. VRR* for two very different [*SP*]

It's a batch !

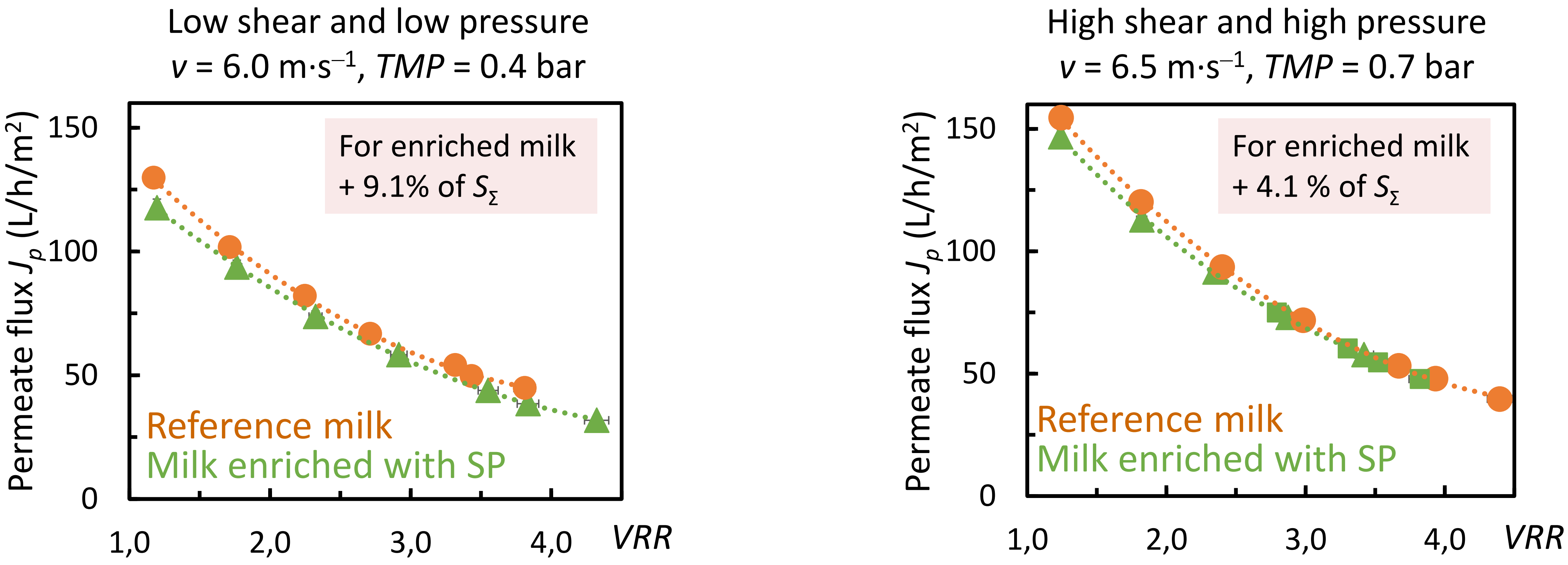


Experiment design & protocol

- *VRR* increased in steps from 1 to 4.2
- Step duration 30 min (tested up to 4h)
- Separate experiments with different combinations of
  TMP = 0.4, 0.7 and 1.0 bar
  with *v* = 6.0, 6.5 and 7.0 m/s
- Analysis of [*CN*] and [*SP*] in permeates and retentates
- Determination of
  $J_p = f(VRR)$
  $[SP]_r = f(VRR)$
  $TR = f(VRR)$
  for reference milk and milk enriched with SP

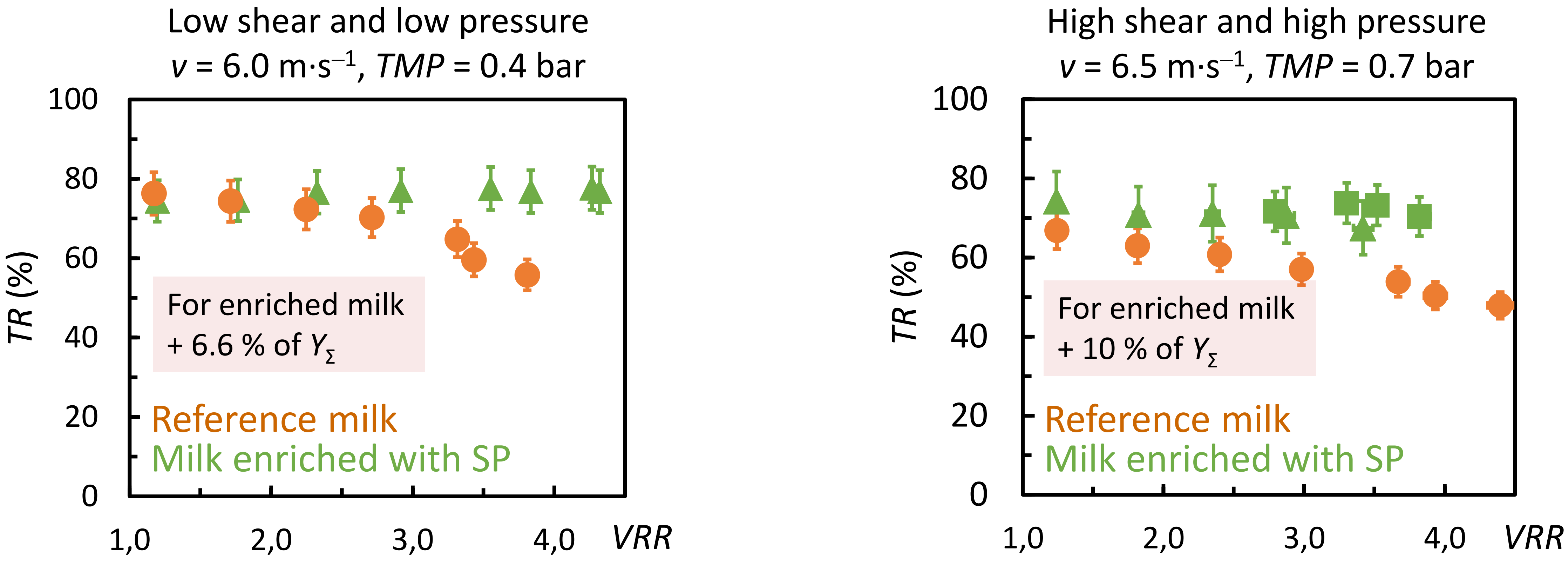


Results : J_p = f(*VRR*)- Permeate flux for reference milk and milk enriched with SP



- Increase of retentate crossflow velocity *v* and *TMP* expectedly increase J_p
- $J_p(VRR)$ of reference milk > $J_p(VRR)$ of milk enriched with SP
- For enriched milk + 9.1% of S_Σ is required at low shear/low pressure and + 4.1% of S_Σ is required at high shear/high pressure (Hyp: 3 stages, VRR_{out}=3.5, Q_{in} = 1 m^3/h)

Results: TR=f(VRR)- Transmission of serum proteins for reference milk and milk enriched with SP



- At $VRR \leq 2.5$, $TR(VRR)$ of reference milk ≈ $TR(VRR)$ of milk enriched with SP
- At $VRR > 2.5$, $TR(VRR)$ of reference milk strongly decreases

  Increase of $[SP]_r$ with VRR in reference milk did compensate the decrease of TR with increase of VRR
- TR expectedly decreased with increase of TMP despite of increase of v
- For enriched milk + 6.6 % of Y_Σ is obtained at low shear/low pressure and + 10% of Y_Σ at high shear/high pressure

  For reference milk, gain in S_Σ costs lost in Y_Σ

Summary

- A methodology is proposed to design the multi-stage separation-concentration system
  → Theoretical approach (mass balance) + empirical data

- *N*-stage system requires less total membrane surface area S_Σ than one-stage system and depends on $J_p(VRR)$
  Values of VRR_i required to minimize S_Σ are calculated as

$$S_{\Sigma,min} = \min_{VRR_i} \sum_{i=1}^{N-1} \left(\frac{1}{J_{p,i}} \left(\frac{1}{VRR_{i-1}} - \frac{1}{VRR_i} \right) \right)$$

- Total yield of permeable component in permeate, Y_Σ depends on $TR(VRR)$ and all VRR_i
  Values of VRR_i required for maximal Y_Σ are calculated as

$$Y_{\Sigma,max} = \max_{VRR_i} \sum_{i=1}^{N} \left(\frac{1}{VRR_{i-1}} \prod_{j=0}^{i-1} b_j - \frac{1}{VRR_i} \prod_{j=0}^{i} b_j \right) \quad \text{where} \quad b_j = \frac{VRR_j}{TR_j(VRR_j - VRR_{j-1}) + VRR_{j-1}}$$

- Values of VRR_i required for maximal Y_Σ can be different from that required for minimal S_Σ (compromise)

- Possible influence of a permeable component concentration in retentate $[SP]_r$ on $J_p(VRR)$ and $TR(VRR)$ must be verified because
  - different combinations of $VRR_{j=1..i}$ can result in different $[SP]_{r,i}$ at same VRR_i
  - batch mode (used for empirical data determination) vs. continuous mode have different $[SP]_r$ at same *VRR*

Thank you for your attention

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