Effect of chitosan based Al species on coagulation performance and ultrafiltration characteristics under different pH conditions

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Shortages of coagulation :

- Low dissolved organic matters removals
- High chemical consumption in eutrophic water treatment
- Low fatal microorganisms removals

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Shortages of ultrafiltration:

Ultrafiltration（UF）has been known to be effective for the removal of

- Not effective in dissolved organic matter removal
- Membrane foulings

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- ✓ High NOM removal efficiency
- ✓ Prevent membrane foulings

Aluminum coagulants

As aluminum salts dosed into water, there would be a series of hydrolysis reactions and different aluminum species generated: Al_a Al_b and Al_c.

Monomeric (Al_a),

Al^{3+}, $Al(OH)^{2+}$, $Al(OH)_2^{+}$

Tridecamer (Al_b),

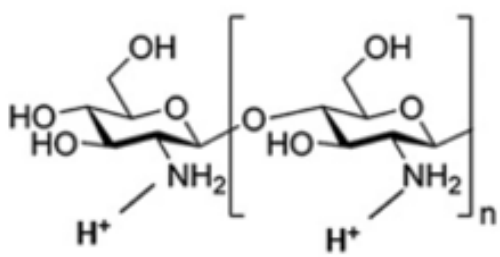
$AlO_4Al_{12}(OH)_{24}(H_20)_{12}^{7+}$

Larger polynuclears (Al_c),

$(AlO_4)_2Al_{28}(OH)_{56}(H_2O)_{26}^{18+}$

Coagulant aid

Chitosan (CS) was the second most abundant biopolymer after cellulose in the word which produced by the deacetylation of chitin.



CS after acid protonation

- **Macromolecular ploymers**
- **Positive bridging effect role**
- **Low cost and non-toxic**

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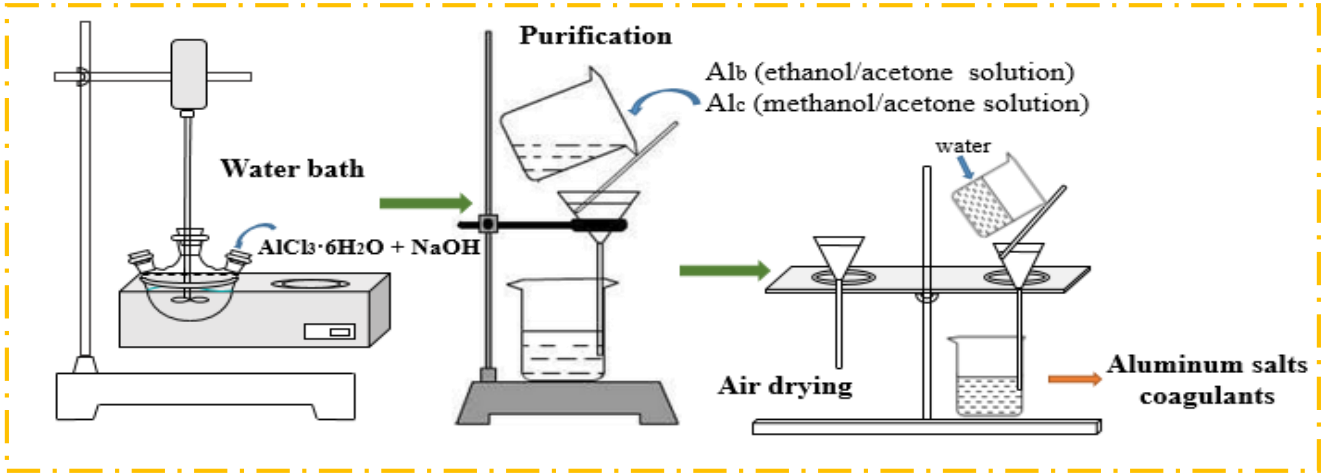
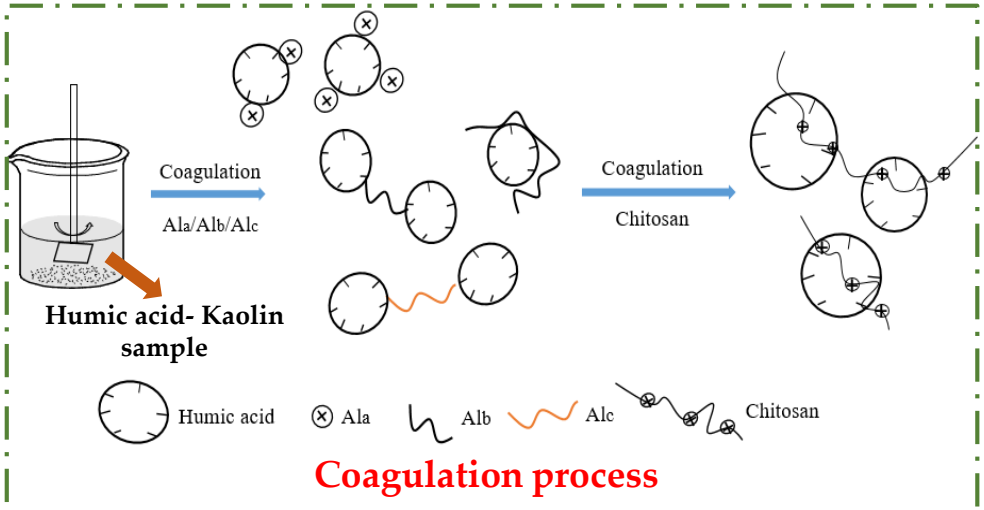
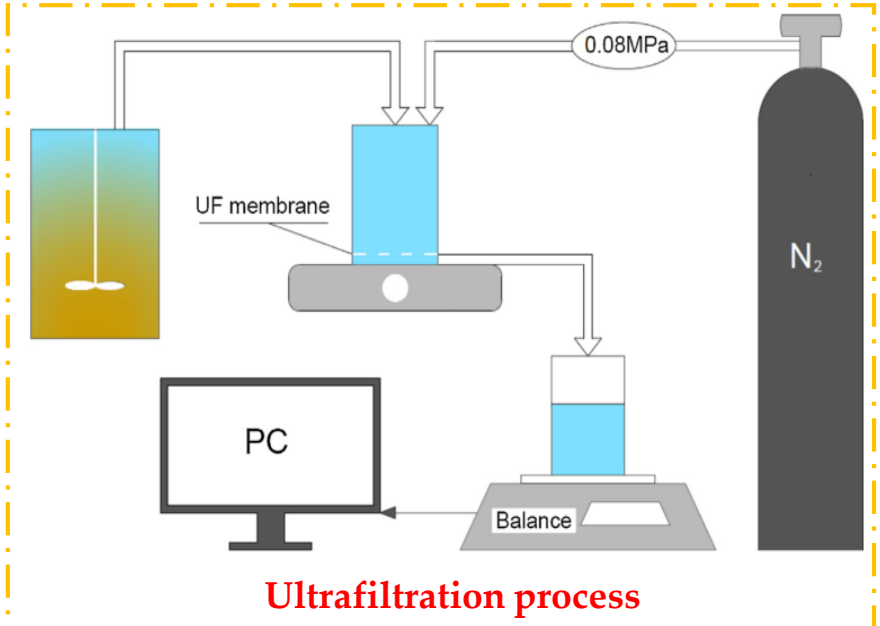




Coagulation process



Ultrafiltration process

Rapid mixing 1.5 min

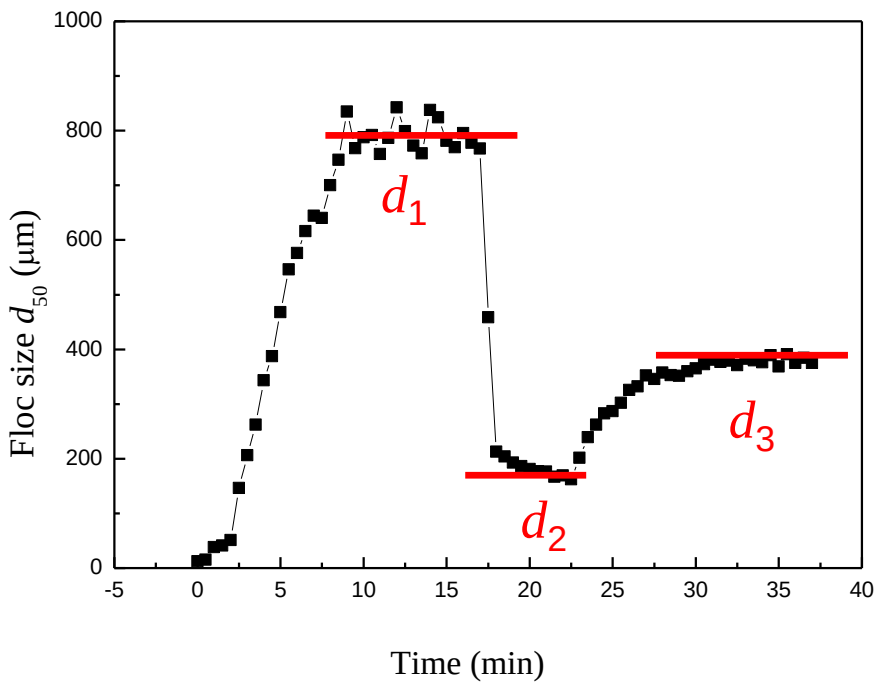
Slow mixing 15 min

Sedimentation 30 min

Collected water samples (Turbidity, UV_{254}, DOC, Zeta potential)

Floc characteristic

- **Floc size (d_{50})**
- **Strength factor (S_f)**
- **Recovery factor (R_f)**
- **Fractal dimension (D_f)**

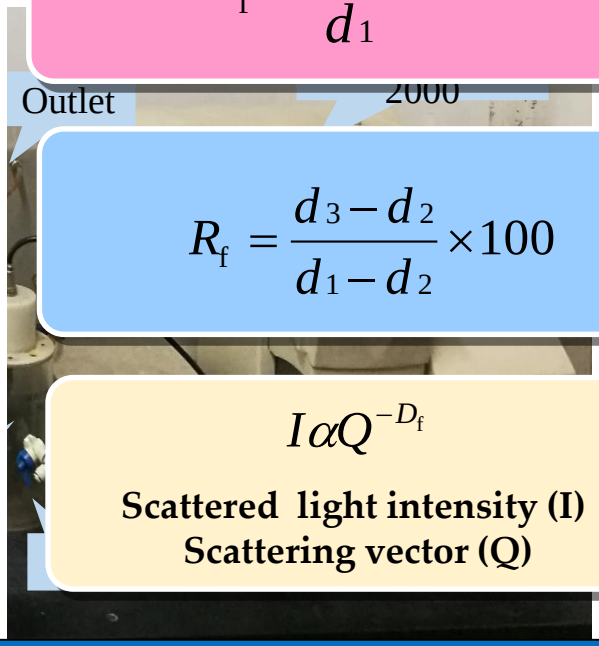


$$S_f = \frac{d_2}{d_1} \times 100$$

$$R_f = \frac{d_3 - d_2}{d_1 - d_2} \times 100$$

$$I \alpha Q^{-D_f}$$

Scattered light intensity (I)
Scattering vector (Q)



- When pre-formed flocs (d_1) are subjected to an increased shear rate, floc breakage occurred.
- The breakage flocs (d_2) could regrow when the shear force disappeared, which achieved a balance (d_3).

Membrane resistances

Total hydraulic resistance (R_t)

External fouling (R_{ef})

- Loosely-attached resistance ($R_{ef\text{-}l}$)
- Strongly-attached resistance ($R_{ef\text{-}s}$)

Internal fouling (R_{if})

- Reversible resistance ($R_{if\text{-}r}$)
- Physically irreversible fouling resistance (R_{irr})

$$R_t = \mathrm{TMP}/(\mu J) = R_m + R_{ef} + R_{if}$$

$$R_{ef-l} = \mathrm{TMP}/(\mu J_1) - \mathrm{TMP}/(\mu J_2)$$

$$R_{ef-s} = \mathrm{TMP}/(\mu J_2) - \mathrm{TMP}/(\mu J_3)$$

$$R_m = \mathrm{TMP}/(\mu J_0)$$

$$R_{irr} = \mathrm{TMP}/(\mu J_4) - \mathrm{TMP}/(\mu J_0)$$

$$R_{if-r} = \mathrm{TMP}/(\mu J_3) - \mathrm{TMP}/(\mu J_4)$$

R_m = clean membrane resistance (m^{-1})
μ = dynamic viscosity of the feed water (Pa·s)

TMP = trans - membrane pressure (Pa)
J/ J_0/ J_1/ J_2/ J_3/ J_4 = permeate flux

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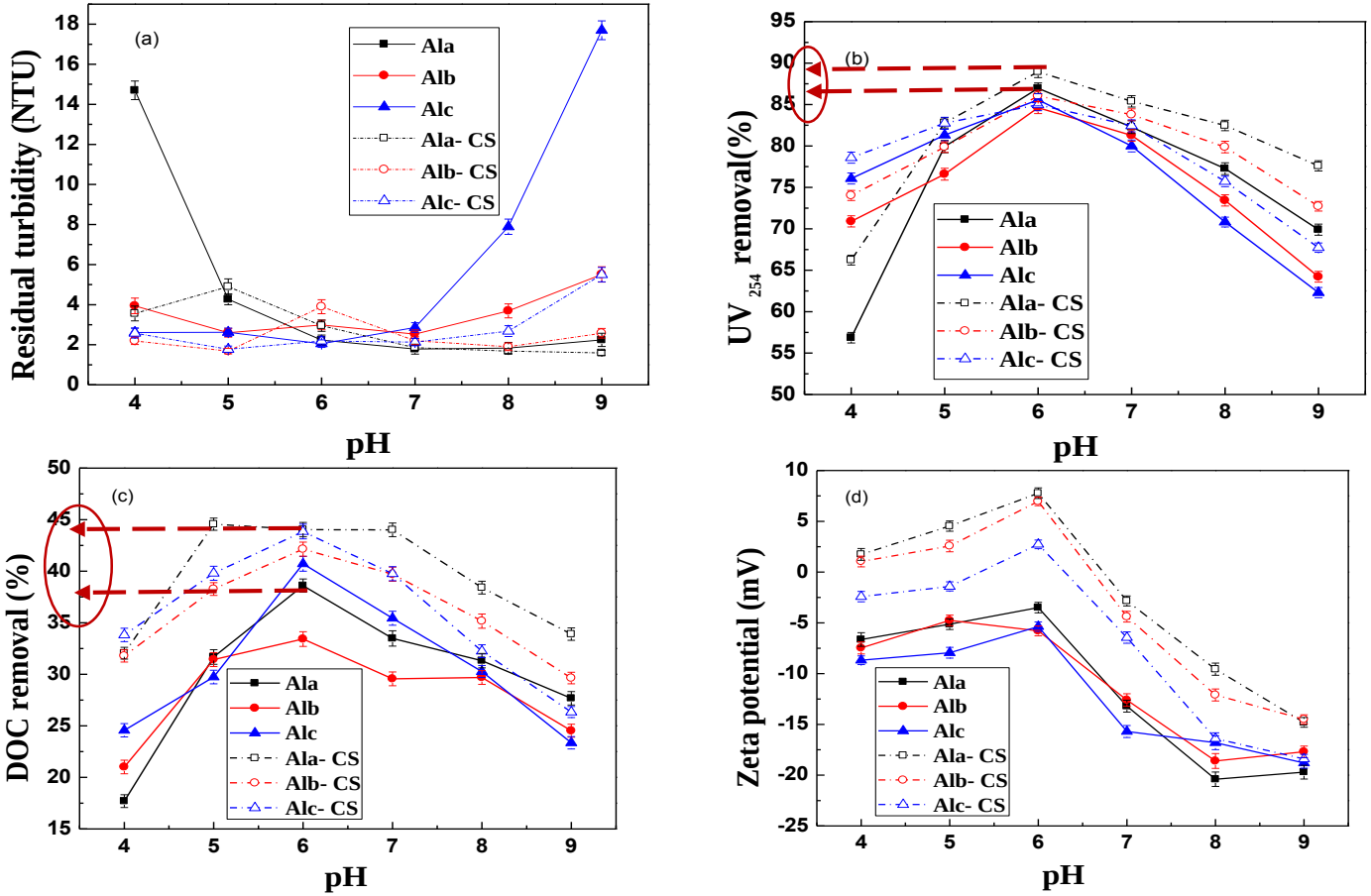
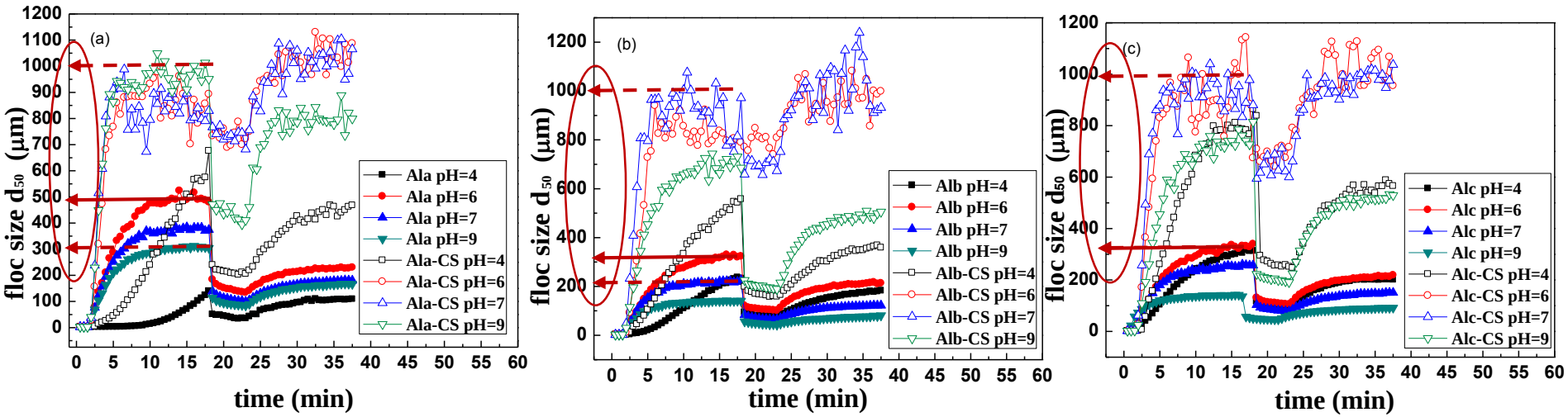
Coagulation performance



- The aluminum dosage of 4 milligrams per liter and chitosan dosage of 1 milligram per liter was selected. The potential dosages (Wang, Yu, et al. Preliminary characterization and application of dual coagulants with the pH range and different Al species in coagulation and ultrafiltration process. Journal of Environmental Sciences, 2016)
- Coagulation efficiency of Al_a/Al_b/Al_c-CS presented a similar trend as Al species, and addition of CS could improve the coagulation efficiency within the pH range.

Floc sizes



- Floc size achieved maximum value at pH 6 by Al species individually, and the floc sizes were smallest at pH 9 except for Al_a.

- ✓ Floc size in steady-state was apparently increased due to the addition of CS.
- ✓ CS as coagulant aid was less affected by pH in water treatment, which indicated that combined Al with CS have a wide pH ranges.

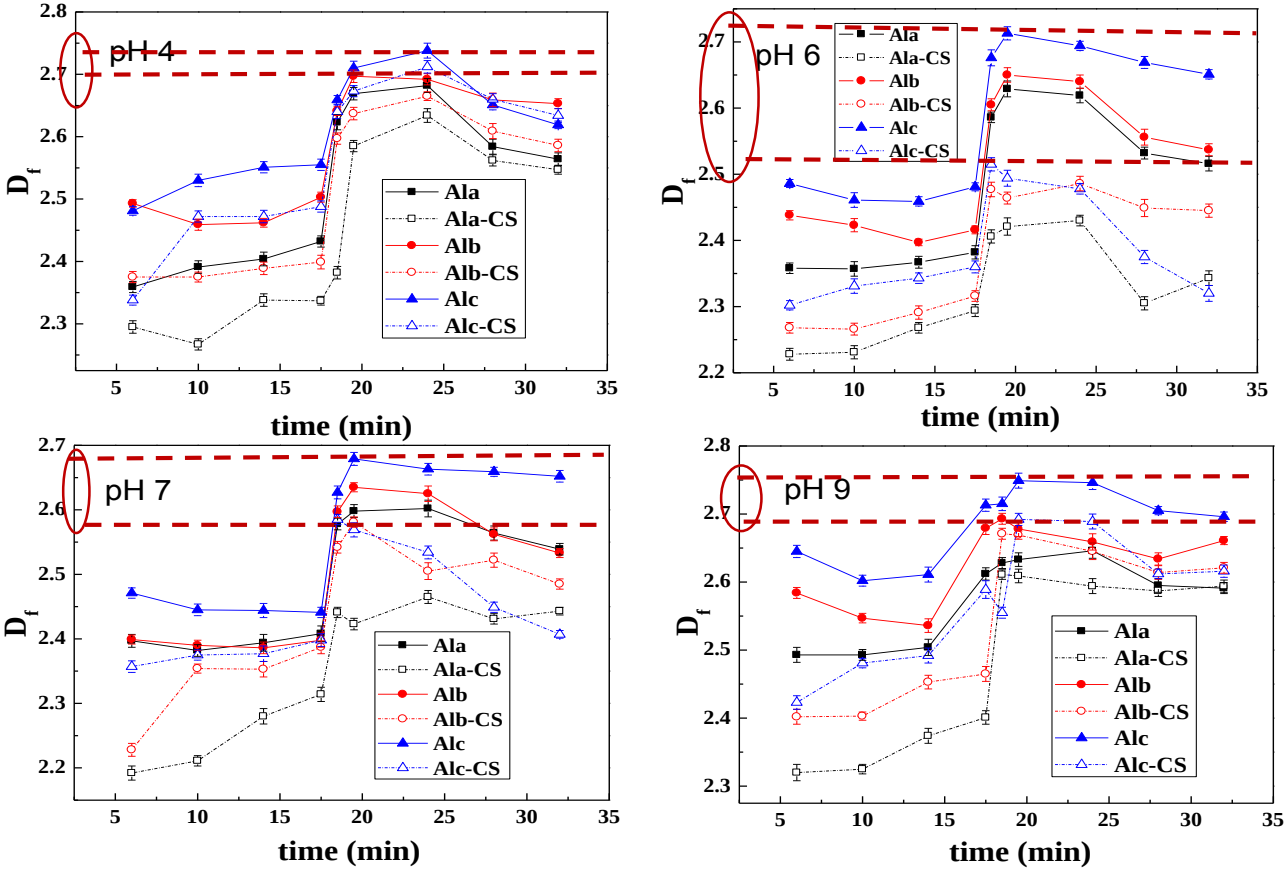
Floc breakage and re-growth

Parameter	Coagulant	pH			
		4	6	7	9
S_f	Al_a	25.14±0.22	27.88±0.35	26.68±0.33	27.55±0.27
	Al_a-CS	30.41±0.27	80.81±0.52	62.20±0.47	23.21±0.25
	Al_b	32.70±0.31	33.41±0.41	30.52±0.36	31.10±0.29
	Al_b-CS	29.48±0.29	92.54±0.57	69.29±0.45	27.57±0.23
	Al_c	30.01±0.33	31.12±0.39	28.55±0.28	30.31±0.25
	Al_c-CS	28.10±0.18	80.98±0.54	68.15±0.39	33.46±0.31
R_f	Al_a	70.33±0.51	67.75±0.45	49.35±0.32	57.40±0.41
	Al_a-CS	55.71±0.47	213.13±0.97	259.35±1.09	71.95±0.45
	Al_b	67.75±0.54	50.34±0.39	35.66±0.28	49.12±0.38
	Al_b-CS	50.21±0.42	133.99±0.75	86.61±0.54	63.71±0.49
	Al_c	48.89±0.39	48.26±0.35	40.52±0.34	50.52±0.35
	Al_c-CS	57.26±0.48	142.09±0.72	155.88±0.77	54.26±0.39

- The S_f and R_f values of floc increased due to the CS addition.

- Maximum S_f and R_f were mainly acquired at pH 6 due to the strongest charge neutralization effect.

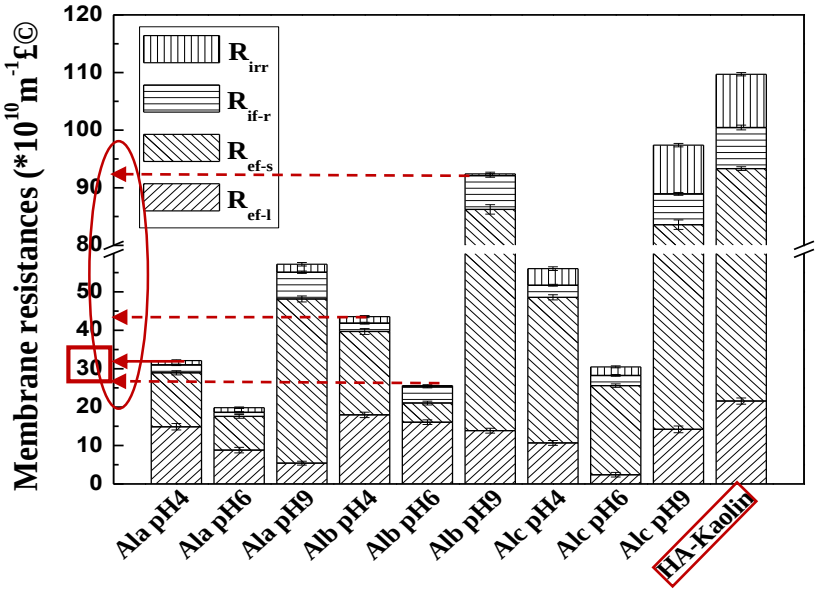
Floc fractal structure

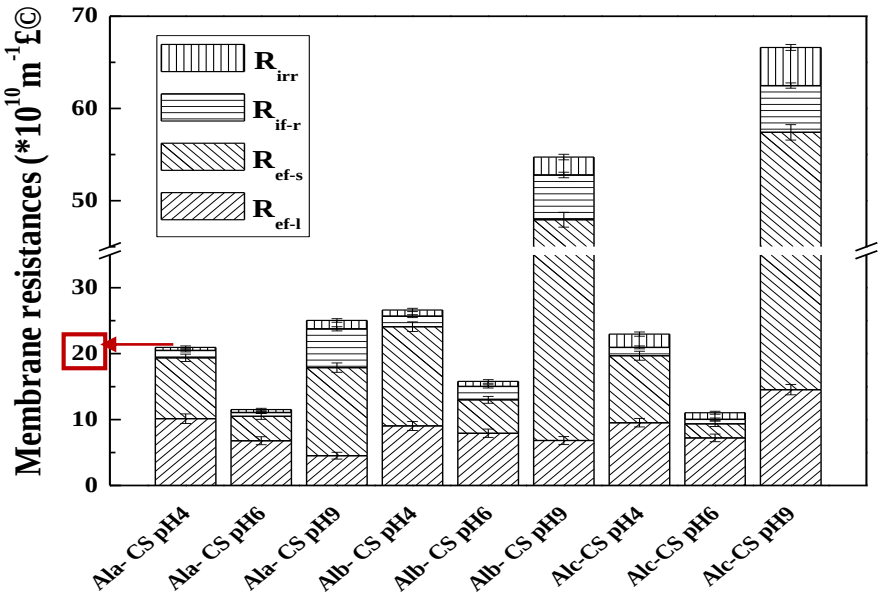


D_f was in a stable state at growth period and regrowth period, but a rapidly increasing was found during the breakage period.

D_f showed a reduction when CS as coagulation aid added.

Membrane resistances





- The most serious membrane resistances was observed when no coagulant was used.
- The reduction of R_t was due to the larger floc sizes and looser structure when CS was added at the same pH.
- In the same system, the smallest total resistances occurred at pH 6, but the maximum value of total resistances achieved at pH 9.

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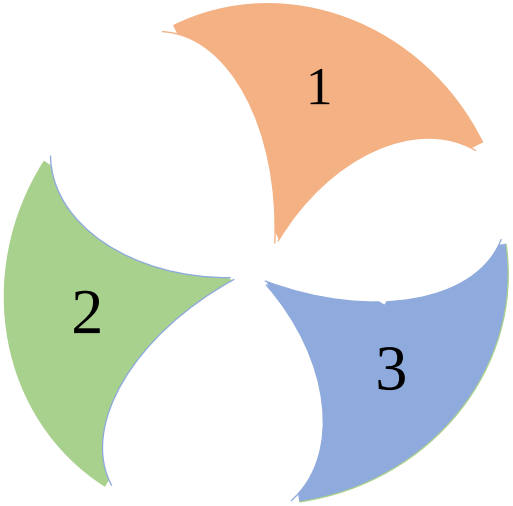
CONCLUSIONS

- HA removal efficiency was the highest at pH 6, and $Al_a/Al_b/Al_c$-CS increased the removal ratios compared with $Al_a/Al_b/Al_c$.

- Flocs under acidic conditions were smaller than these under alkaline conditions except for Al_c. Moreover, larger and looser flocs were formed when CS dosed.



- Membrane resistances were related to membrane fouling which impact on floc size and structure.

Thank you !