# Supporting Information

## Dual-functional ice/water interface allows high-yield formation of $Al_{13}$ with low energy

Liang Kuang[1,3], Ning Li[2,3], William A.Jefferson[1,], Yaohui Bai[1,3], Qinghua Ji[1,3] , Huijuan Liu[2,3], and Jiuhui Qu[*1,3]

[1] Key Laboratory of Drinking Water Science and Technology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing, 100085, China

[2] State Key Laboratory of Environmental Aquatic Chemistry, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing, 100085, China

[3] University of Chinese Academy of Sciences, Beijing, 100049, China

*Corresponding author:

Telephone/Fax: +86-10-62849160
E-mail: jhqu@rcees.ac.cn

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## S.1 Materials

All chemicals were reagent grade or higher. Millipore water(18.2MΩ) was used in the preparations of all solutions. Commercial PACl (Tianjin, China) was a commercial PACl product containing 28% of $Al_2O_3$, the content of $Al_b$ in which was 33% by Ferron method.

## S.2 Experimental Methods

### 2.1 Characterization

The Ferron method was used for the characterization of aluminium in synthetic product.[1]The Ferron colorimetric method is based on the determination of the absorbance at 370nm.There are three fractions which could be operationally considered as $Al_a$, $Al_b$, and $Al_c$, corresponding to monomeric , medium polymer and larger polymer species or $Al(OH)_3$， respectively. According to literature[1], the absorbance detected at 1min represented for $Al_a$, and which was detected at 120min represented for $Al_a$ + $Al_b$, thus $Al_c$ was obtained by $Al_T$ minus $Al_a$ and $Al_b$. Many investigations regarded the $Al_b$ species as the $Al_{13}$ species

## S.3 Figures and tables

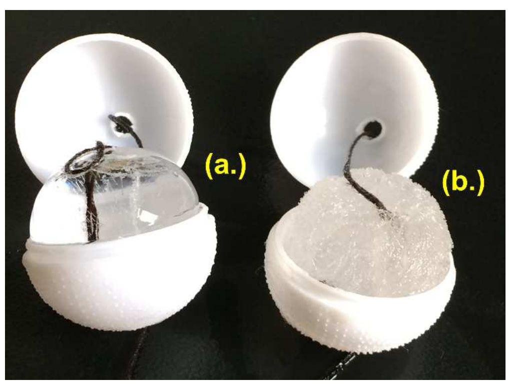


**Figure S1.** Comparison of images of the ice ball(water) towards the ice ball (NaOH solution) (a). The ice ball is frozen from Millipore water. (b) The ice ball is frozen from NaOH solution.

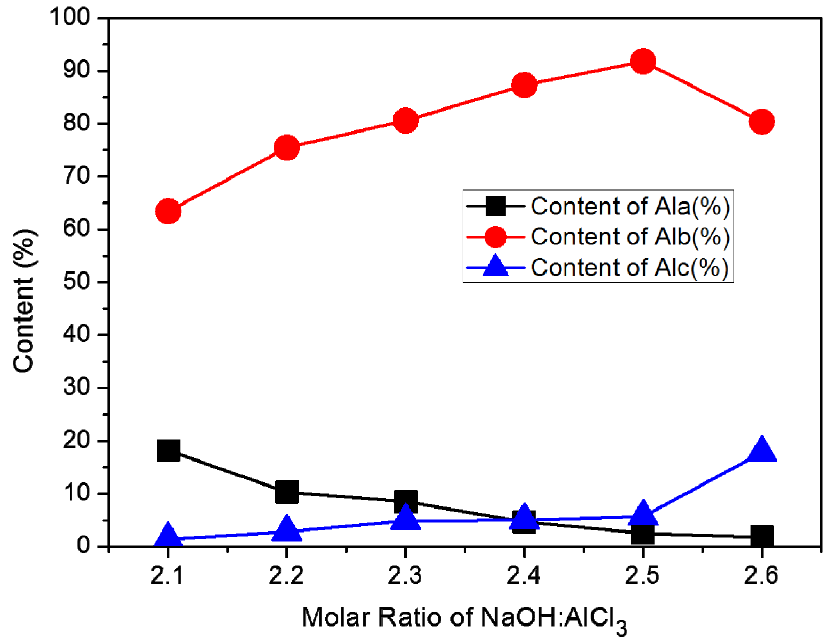


**Figure S2.** The characterization of synthetic product by Ferron method at different molar ratios of NaOH: $AlCl_3$

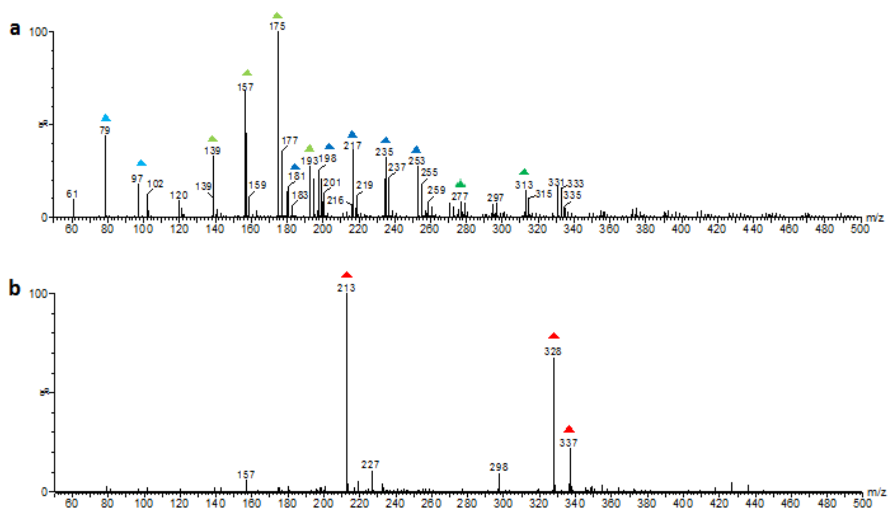


**Figure S3.** ESI-TOF-MS spectra of AlCl3 and the synthetic product (0.05mol/L). (a), the $AlCl_3$ solution as a raw material. (b), the synthetic product. The ESI-TOF-MS conditions refer to the literature[2], the parameters were: capillary voltage 3500.0 V, sample cone voltage 70 V, extraction cone voltage 5 V, source temperature 120 ℃, desolation temperature 150 ℃, cone gas ($N_2$) flow rate 300 L $h^{-1}$, and mass range 50~1000.

（▲ $Al_1$, ▲ $Al_2$, ▲ $Al_3$, ▲ $Al_4$, ▲$Al_{13}$）

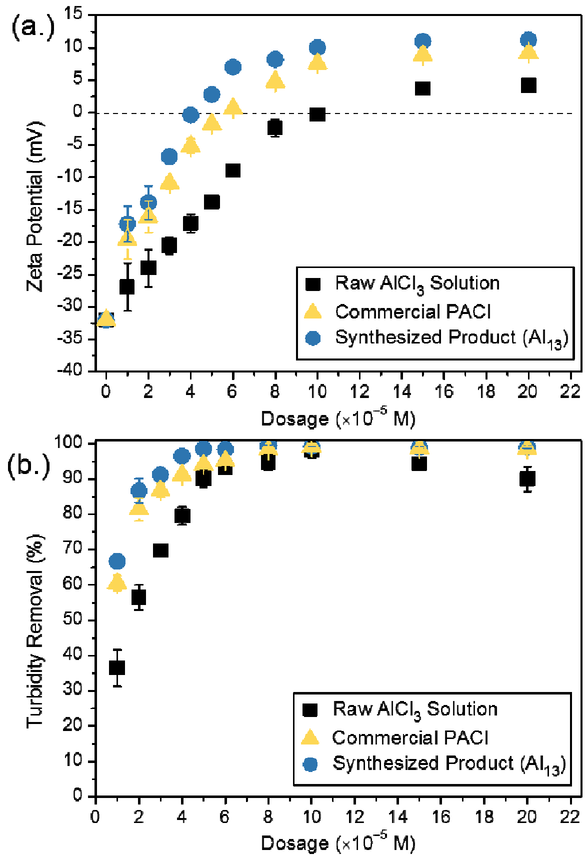


**Figure S4**. The Zeta potentials and turbidity changes of coagulated material as a function of coagulant dose. (The initial turbidity was 70NTU, the initial pH was 7 and the ionic strength was $10^{-3}$mol/L)

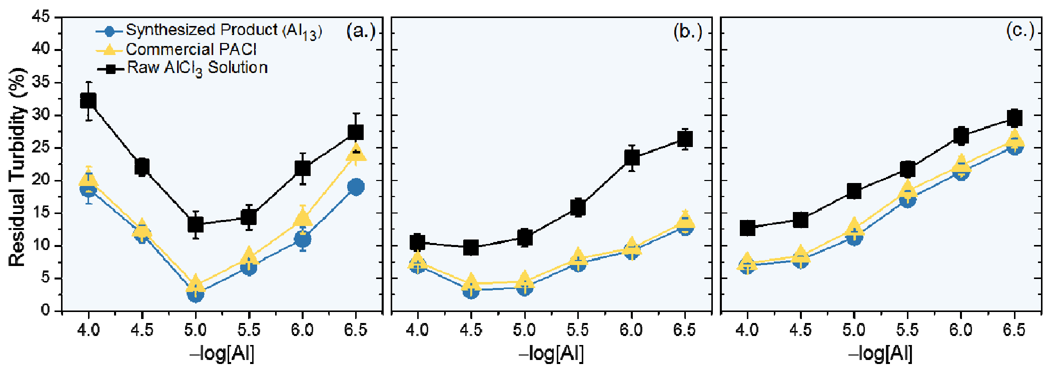


**Figure S5.** The removal of turbidity under different initial pH (a)pH=5 (b) pH=7(c)pH=9.

**Table S1**. Comparison of experimental m/z with theoretical m/z of different aluminium species

| m/z | Aluminium species formula | Experimental m/z | Theoretical m/z |
|---|---|---|---|
| 79 | $Al(OH)_2(H_2O)^+$ | 79.01 | 79.01 |
| 97 | $Al(OH)_2(H_2O)_2^+$ | 97.03 | 97.03 |
| 139 | $Al_2O_2(OH)(H_2O)_2^+$ | 139.00 | 139.00 |
| 157 | $Al_2O_2(OH)(H_2O)_3^+$ | 157.01 | 157.01(157.0151) |
| 175 | $Al_2O_2(OH)(H_2O)_4^+$ | 175.03 | 175.03 |
| 193 | $Al_2O_2(OH)(H_2O)_5^+$ | 193.05 | 193.05 |
| 181 | $Al_3O_4(H_2O)_2^+$ | 180.97 | 180.97 |
| 199 | $Al_3O_4(H_2O)_3^+$ | 198.99 | 198.99 |
| 217 | $Al_3O_4(H_2O)_4^+$ | 217.00 | 217.00 |
| 235 | $Al_3O_4(H_2O)_5^+$ | 235.02 | 235.02 |
| 253 | $Al_3O_4(H_2O)_6^+$ | 253.03 | 253.03 |
| 277 | $Al_4O_5(OH)(H_2O)_4^+$ | 276.99 | 276.99 |
| 313 | $Al_4O_5(OH)(H_2O)_6^+$ | 313.02 | 313.02 |
| 213 | $AlO_4Al_{12}O_{14}^{3+}$ | 212.92 | 212.92 |
| 328 | $AlO_4Al_{12}O_{14}(OH)^{2+}$ | 327.88 | 327.88 |
| 337 | $AlO_4Al_{12}O_{13}(OH)_3^{2+}$ | 336.89 | 336.89 |

**Table S2**. Comparison of various synthetic methods used to prepare $Al_{13}$

| Preparation Method | Energy needs for | Required External input | Waste streams | Content (%) | Time Consuming | Shelf-life[a] | Ref. |
|---|---|---|---|---|---|---|---|
| Micro-injection | vigorous stirring | | none | 44（$Al_T^b$=0.2M） | hrs | Several Months | **3** |
| Furrier Method | pump | Electricity | Spent marble granulate, $Al(OH)_3$ by-product | 93($Al_T$=5.29*10$^{-3}$) | 1 hr or more | Several Months | **4** |
| Electrolysis | electrode | Electricity | Anode mud | 89（$Al_T$=0.2M） | hrs | Several Months | **3** |
| Membrane Reator | Membrane pump | Electricity | Clogging and back-wash waste | 79($Al_T$=0.27M) | | Several Months | **5** |
| Ice-melting | Freezing (or cold climate) | none | none | 90($Al_T$=0.2M) | mins | Over 6months (up to now) | |

**a In the other methods, in exact, the shelf-life is not given in the literature, but it can be assumed to be at least several months.**

**b.The $Al_T$ here refers to the total aluminum in the synthetic product.**

## S.4 Results and discussion

Set the integrated area of Na aluminate in Fig.2 to 1, then the area at 0ppm was 11.32(Fig.2a), the area at 0ppm and 63ppm was 0.49 and 0.8, respectively. (Fig.2b) If we multiplied the signal of 63ppm by 13, the result is about 90% of the 0ppm signal of Fig.2a. This calculation is consistent with the result from the $Al_{13}$ concentration divided by the $Al_T$ determined from ICP-OES.

## References


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