Online Supporting Information for

# "Green Production of Ultrahigh-Basicity Polyaluminum Salts with Maximum Atomic Economy by Ultrafiltration and Electrodialysis with Bipolar Membranes"

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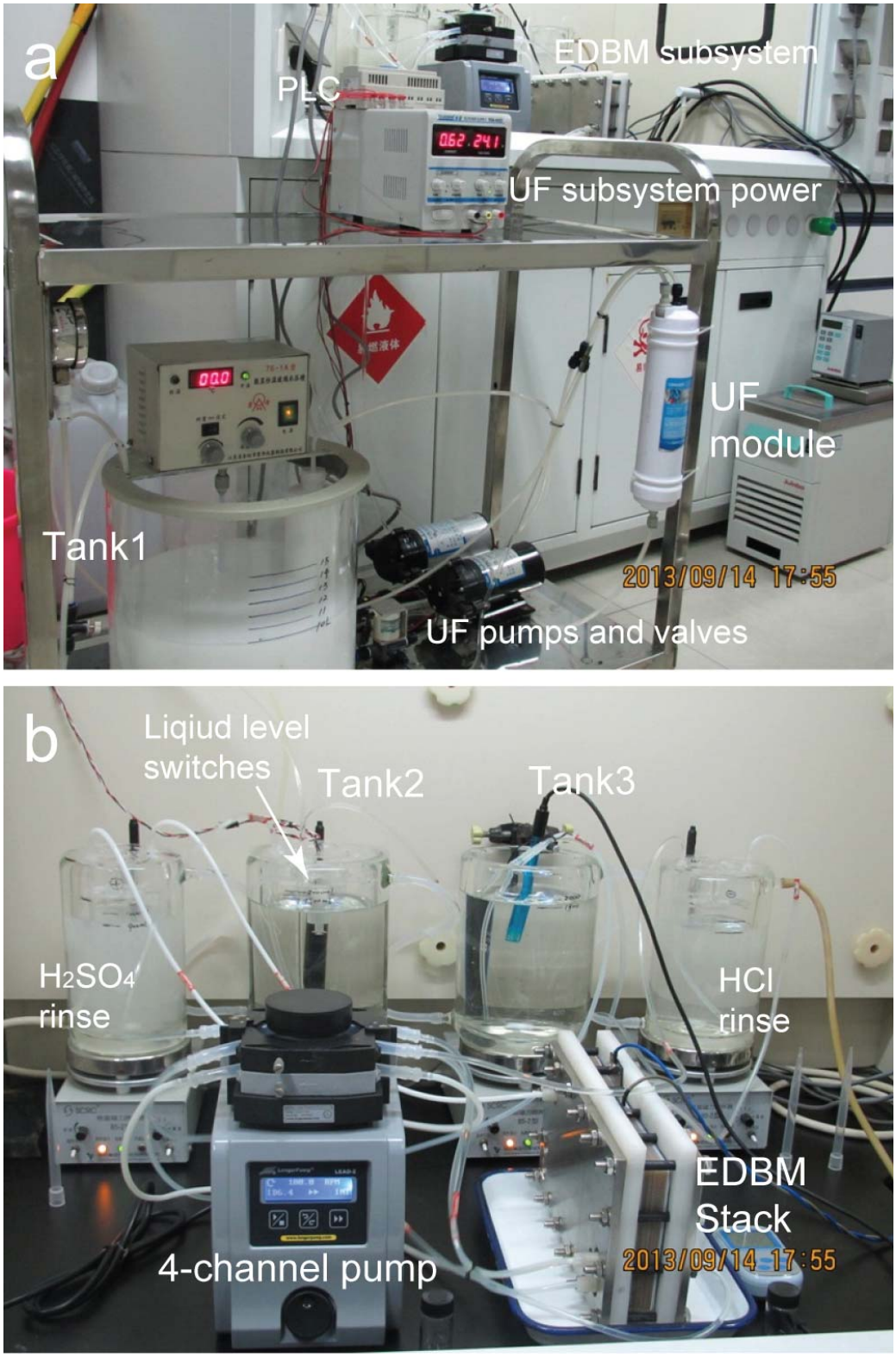


**Figure S1.** Photographs of the integrated system. The UF subsystem (a) and the EDBM subsystem (b).

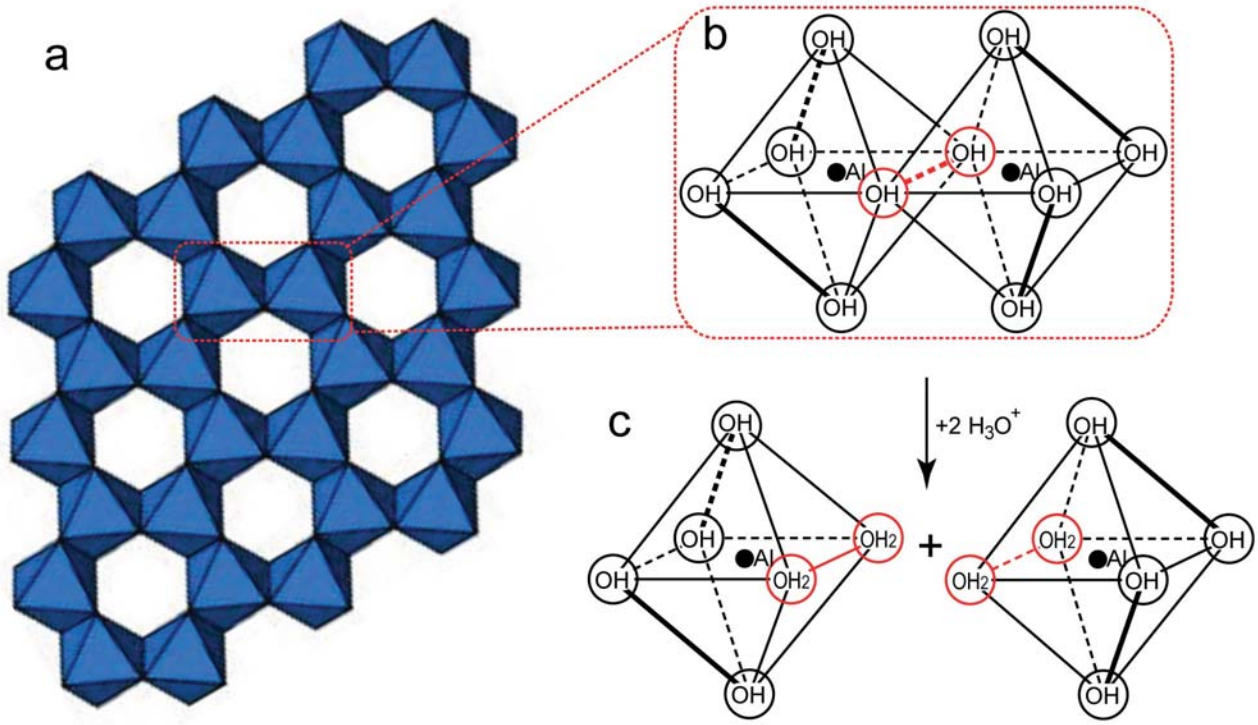


**Figure S2.** Solution chemistry of $Al(OH)_3$ reacting with free acids. (a) Schematic layer structure of aluminum hydroxide (gibbsite). Each octahedron represents 1 $Al^{3+}$ surrounded by 6 $OH^-$, and each $OH^-$ is shared by 2 adjacent octahedrons, giving an empirical formula of $Al(OH)_3$. (b) A close look of the adjacent aluminum octahedrons. The shared double hydroxide bridges are emphasized as thick lines. (c) The combined hydroxide bridges are taken apart when attacked by hydrated protons. Separated $[Al(H_2O)_6]^{3+}$ octahedrons are the final products when gibbsite reacts with adequate free acids.

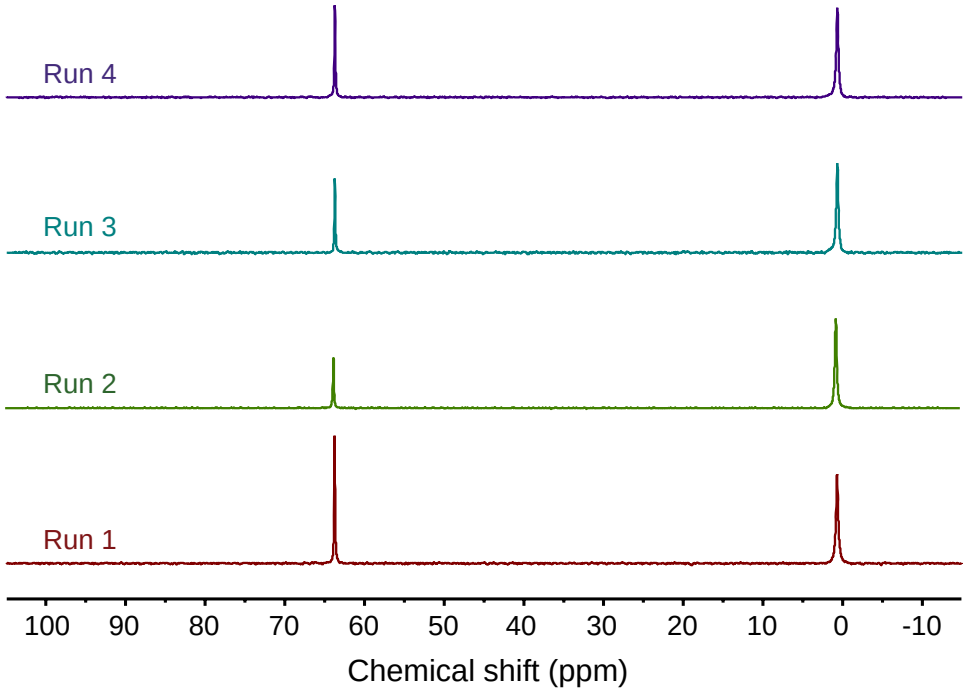


**Figure S3.** $^{27}$Al-NMR spectrum of the final polyaluminum products in all batches.

| Items | UF+EDBM process | | Notes |
|---|---|---|---|
| **Lab system parameters** | | | |
| System production cycle | 2.0 | hour/batch | *EDBM with large mixing tank, including 30 min clean* |
| Final product basicity | 70% | | *See Fig. 4c for details* |
| Polyaluminum salt yield | 0.90 | mol/batch | *Calculated as Al* |
| | 0.101 | kg/batch | *Calculated as* $Al_2(OH)_{4.2}Cl_{1.8}\cdot 2H_2O$ *solids* |
| UF effective membrane area | 0.20 | $m^2$ | |
| UF average power | 20.0 | W | *15 W for normal and 40 W for flush operation* |
| UF energy consumption | 0.040 | kWh/batch | |
| EDBM effective membrane area | 0.168 | $m^2$ | *187 cm2 x 9 cell pairs* |
| EDBM energy consumption | 0.250 | kWh/batch | *See Fig. 3b for details* |
| **Full system cost estimation** | | | |
| Scale up factor from lab system | 200 | | *Cost analysis appliable for large system only* |
| UF effective membrane area | 40 | $m^2$ | |
| UF module cost | 1,200 | $ | *Litree LH3-1060-V module (same fiber, 40 m2 area)* |
| EDBM effective membrane area | 33.7 | $m^2$ | |
| EDBM total membrane area | 42.1 | $m^2$ | *80% effective area ratio* |
| BP membrane cost | 30,294 | $ | *$720/m2 for BP membrane* |
| Anion membrane cost | 15,147 | $ | *$360/m2 for anion membrane* |
| EDBM stack cost | 68,162 | $ | *1.5 times for membranes* |
| Cost for core membrane modules | 69,362 | $ | *UF Module + EDBM stack* |
| Peripheral equipment cost | 34,681 | $ | *50% of core membrane modules* |
| Capital cost | 104,042 | $ | |
| Maintenance cost | 10,404 | $/year | *10% of capital cost per year* |
| System lifespan | 2 | year | |
| Available working time | 8,000 | hour/year | |
| Total investment in system lifespan | 124,851 | $ | *Capital + Maintenance* |
| Total yields in system lifespan | 162,216 | kg | *as* $Al_2(OH)_{4.2}Cl_{1.8}\cdot 2H_2O$ *solids* |
| Capital cost | 0.770 | $/kg | |
| Energy consumption | 2.86 | kWh/kg | *UF + EDBM* |
| Electricity charge | 0.10 | $/kWh | |
| Energy cost | 0.286 | $/kg | |
| Total cost (exclude feed chemicals) | 1.06 | $/kg | *as* $Al_2(OH)_{4.2}Cl_{1.8}\cdot 2H_2O$ *solids* |

**Table S1.** Cost estimation of the integrated process.