Supporting Information

Aggregation and deposition of *in-situ* formed magnesium hydroxide in the presence

of hydrolyzed polyacrylamide

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Experimental Section

Supporting Data:

Figure S1: Simulation results of proportions for each components in IFM.

Figure S2: Electrophoretic mobilities of IFM and precipitates aged after 6 days as a

function of pH.

Figure S3: Zimm plot of HPAM-1, HPAM-2 and HPAM-3.

Table S1: Weight-average molecular weight and hydrolysis degree of three kinds of

HPAM with other static light scattering parameters.

Detailed procedure of adsorption Experiments

For surface precipitation, according to the order of adding $MgCl_2$, two different methods were involved: (1) $MgCl_2$ with a predetermined amount was dropwise added into the HPAM solution with preset pH 12 under rapid mix (300 rpm) in a glass beaker under magnetic stirring. When the drop procedure was completed, a slow mix was applied to confirm the homogeneous mix of the mixtures. The concentration of the HPAM solution was 250 ppm, with final pH of 11.70 ± 0.03. (2) NaOH was dropwise added into the HPAM solution which was previously dissolved with a certain amount of $MgCl_2$. The mix procedure was the same as above. The final concentration of HPAM and $MgCl_2$ was the same as above, and the error of pH was within 0.01. For heterocoagulation: (3) in a separate beaker, an equal volume (75 mL) of $MgCl_2$ and NaOH was mixed to generate $Mg(OH)_2$. The beaker containing a desired amount of $Mg(OH)_2$ was then transferred to another beaker containing 150 mL HPAM solution. A slow mix was applied after a rapid mix, and sustained for 2 min. The final amount of HPAM and $Mg(OH)_2$ was consistent with the above experiments. The final determination of the dosage of each reagent was determined by trial and error according to Visual Minteq 3.0. (4) Preformed $Mg(OH)_2$ aged after 6 d was added into the HPAM solution, and the specific procedure was the same as above. It should be noted that the solution pH of the $Mg(OH)_2$ aged after 6 d was lower than the freshly precipitated $Mg(OH)_2$. So, after the mixture, the pH was adjusted to 11.70 to maintain consistency. $Mg(OH)_2$ formed in the first three conditions can be treated as freshly precipitated magnesium hydroxide, although the former two are *in situ* formed, but the third one is *ex-situ* formed.

Procedure of simulation using Visual Minteq 3.0

A Visual Minteq 3.0 [37] software was used to simulate the solution component during the precipitation process of magnesium hydroxide and the pH changes of the solution.

The software has a huge chemical equilibrium model database and offer multiple options when choosing the solution pH. In our experiments, we set an initial pH (11, 11.5 12, 12.3, 12.5, and 12.8) to make it convenient for the follow-up experiments to be conducted, so the option of calculating pH from solution chemistry conditions was chosen to conduct the simulation. The concentration of the Mg^{2+} was set at 5.3 mM to gain the consistence with the experimental section. A series of pH needed to be simulated in our research, so the "Multi-problem/Sweep" program was used to generate the results. After the calculation, the proportions for each components in IFM were shown in Fig. S1, and the initial-pH and equilibrium-pH relationships were shown in Fig. 4.

Detailed procedure of time-resolved dynamic light scattering

The predetermined volume of HPAM stock solution was first filtered by a 0.45 μm syringe filter (Acrodisc; Pall Corporation) and injected into different vials. The filtered solution was then settled for 1 d to avoid the potential influence of filtration. Then, the $MgCl_2$ solution, after filtering by a 0.2 μm filter (Acrodisc; Pall Corporation), was injected into the HPAM solution. Numerous reports have found that the concentrated divalent cations have a strong influence on the hydration diameters of HPAM [1, 2]. However, under our experiment conditions, the concentration of the Mg^{2+}

was rather low, and we did not observe a change in hydration diameter over a rather long time scale. Nevertheless, to avoid potential influence, we conducted the subsequent experiments after 5 h.

Mixing methods may influence the initial aggregation rate [3], especially for our system, in which the magnesium hydroxides are *in-situ* formed. Two syringes, one containing predetermined amounts of NaOH solution and the other containing HPAM/MgCl$_2$ mixture solution, were simultaneously injected into a vial at a certain speed manually to obtain a mixed solution. The operation time was within 30 s, and the vial was then transferred quickly into the vat of the light scattering system to begin the TR-DLS measurements.

References

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[2] S. Lages, R. Michels, K. Huber, Coil-Collapse and Coil-Aggregation due to the Interaction of Cu2+ and Ca2+ Ions with Anionic Polyacylate Chains in Dilute Solution, Macromolecules, 43 (2010) 3027-3035.

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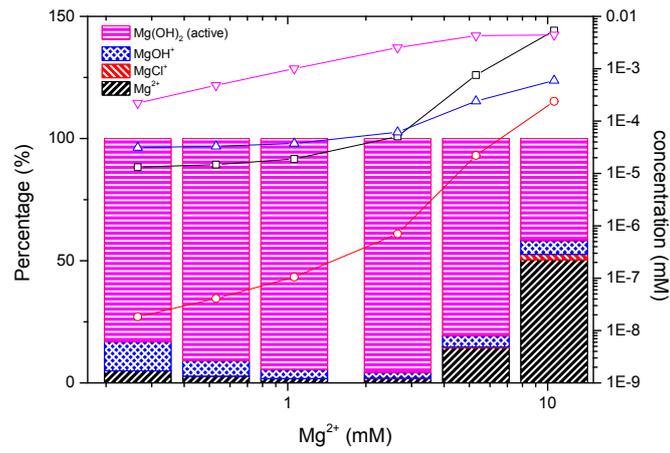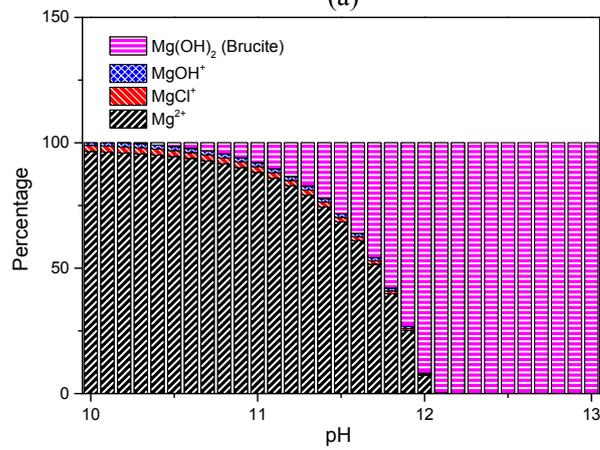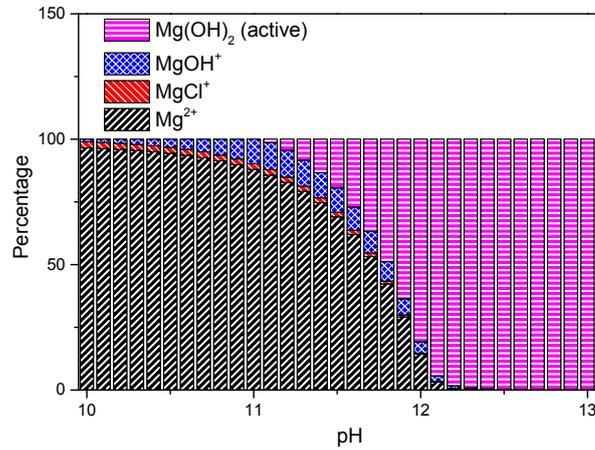
Fig. S1. Simulation results of proportions for each components in IFM. (a) proportions and concentrations of each components as a function of Mg^{2+} concentration. (b) proportions and concentrations of each components as a function of pH, assumed final product was brucite. (c) proportions and concentrations of each components as a function of pH, assumed final product was active $Mg(OH)_2$.
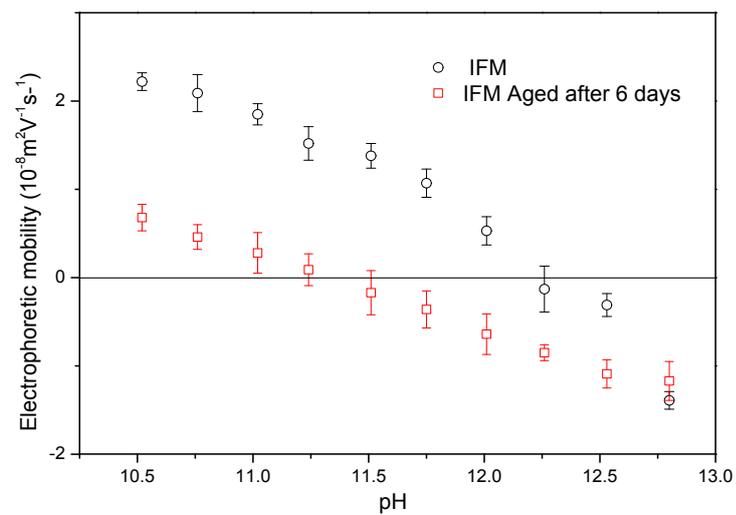
Fig. S2. Electrophoretic mobilities of IFM and precipitates aged after 6 days as a function of pH. The data was measured immediately after the pH was adjusted to desired value. For the precipitates aged after 6 days, the pH was further measured. Scale bars represent the standard deviation.
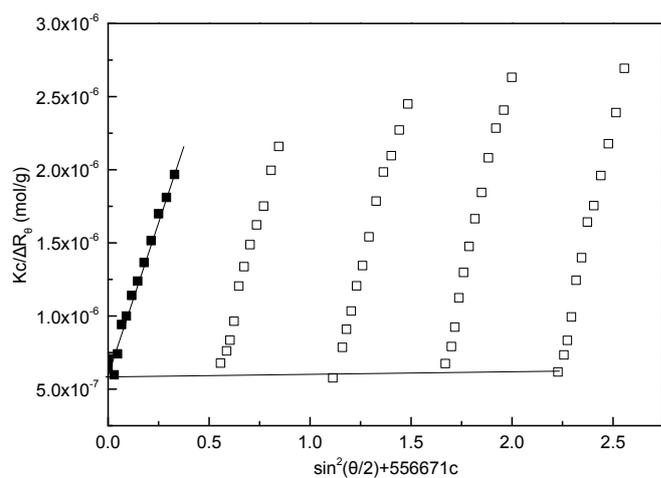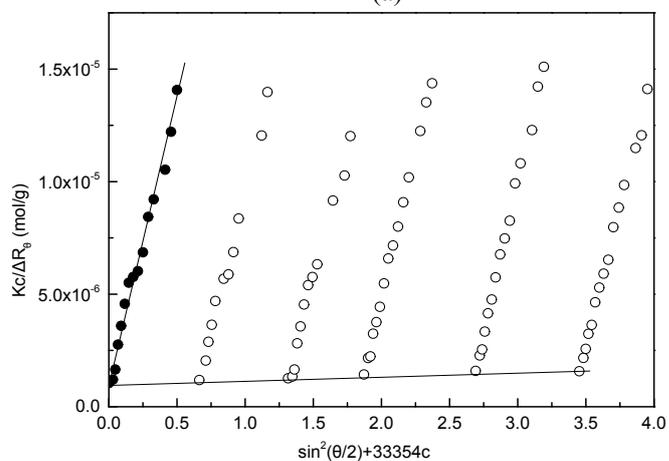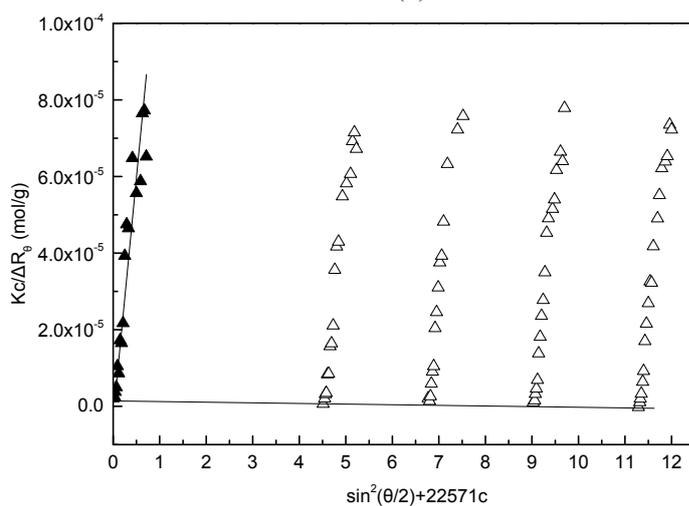
Fig. S3. Zimm plot of (a) HPAM 1, $M_w = 1.52 \times 10^6$ g/mol. (b) HPAM 2, $M_w = 9.14 \times 10^5$ g/mol. (c) HPAM 3, $M_w = 4.70 \times 10^5$ g/mol.

Table S1. Weight-average molecular weight and hydrolysis degree of three kinds of HPAM with other static light scattering parameters.

	$M_w\ (g \cdot mol^{-1})$	$A_2\ (cm^3 \cdot mol \cdot g^{-2})$	$R_g\ (nm)$	Hydrolysis Degree (%)
HPAM 1	1.52×10^6	-4.10×10^{-3}	136.17	25±3
HPAM 2	9.14×10^5	2.63×10^{-3}	122.43	25±1
HPAM 3	4.70×10^5	-1.82×10^{-3}	77.4	24±2