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Electronic Supplementary Information

Controlled exponential growth in layer-by-layer multilayers using high gravity fields

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1. Introduction to high gravity technique.

The high gravity technique was first reported by Prof. Colin Ramshow from Imperial Chemical Industries in 1979. In the high gravity equipment, the solution of building block is continuously pumped into the cavity of a rotator through the liquid inlet pipe and sprayed onto a liquid distributor by a nozzle. Then the sprayed liquid flows through a packing layer (normally porous wire meshes) around the rotator; with the rapid rotating of the high gravity machine, the liquid is remarkably accelerated. At the edge of the packing layer four slits in four directions are set for inserting quartz or silicon substrates (three red or green substrates inserted in three front slits are shown in Fig. 1a-d). When the liquid with high velocity leaves the packing layer by centrifugal force, it flows through the substrates rapidly and the building blocks within solutions are deposited on the substrate. In the above processes, the liquid is highly dispersed by the packing layer, tore and continuously formed renewed interfaces under the field with hundreds to thousands times of gravity acceleration, which improves the mass transfer velocity by one to three magnitude order. In our previous reports, we have demonstrated that the diffusing process of the building blocks could be highly enhanced under this high gravity field and thus contributed to an accelerated surface adsorption. Finally, the liquid is thrown to the cavity and finally leaves the machine through the liquid outlet.

The high gravity technique has been widely used in many fundamental research and industrial fields, such as distillation, adsorption and separation in chemical engineering processes, yeast fermentation, synthesis of nanoparticles. On the aspect of intensified chemical engineering process, high gravity equipment with a diameter of 3 m and a height of 3 m has been used to replace the original reactor with a diameter of 6 m and a height of 30 m.

2. UV-visible spectra of each PAA/PAH-Por bilayer.

The stepwise characterization of UV-visible spectra after deposition of each PAA/PAH-Por bilayer in the absence and in the presence of high gravity field is shown in **Fig. S1** and **Fig. S2**, respectively.

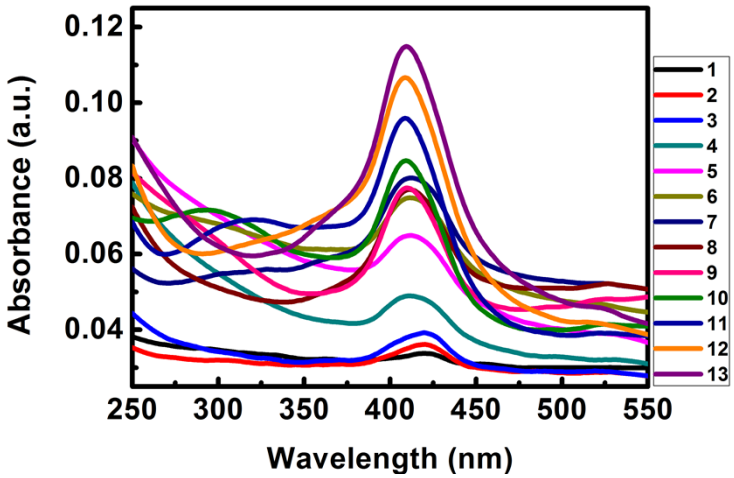

Fig.S1 UV-visible spectra of PAA/PAH–Por multilayer fabricated through LbL assembly under normal dipping conditions.

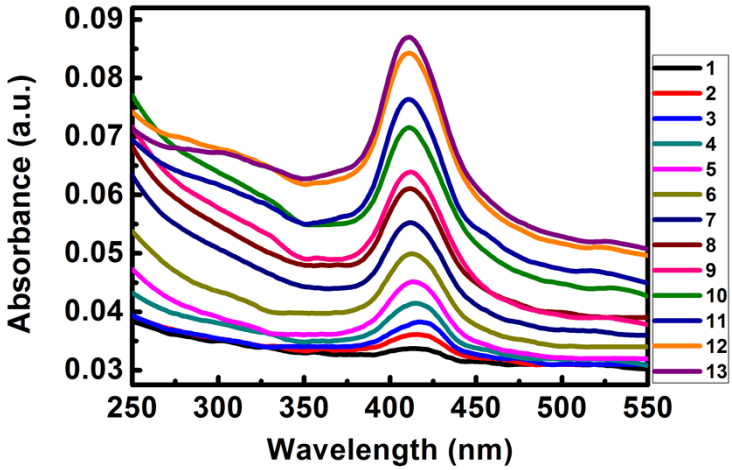

Fig.S2 UV-visible spectra of PAA/PAH–Por multilayer fabricated through LbL assembly under high gravity field.

3. Shear stress produced by high gravity field

For fluids moving along solid boundary, shear stress will occur on the boundary. For a Newtonian fluid, the shear stress (F) is defined as the gradient of fluid velocity (dv/dy), $F=-\mu(dv/dy)$, where μ is the dynamic viscosity of the fluid, v is the fluid velocity along the boundary and y is the height above the boundary as shown below. In the high gravity equipment, the main flow velocity could be accelerated remarkably due to a high centrifugal acceleration whose value is hundreds to thousands times of gravity acceleration. Meanwhile, the velocity of the fluid near the substrate is zero. Therefore, this phenomenon leads to a large gradient of fluid velocity and thus contributes a large shear stress (μ=constant).

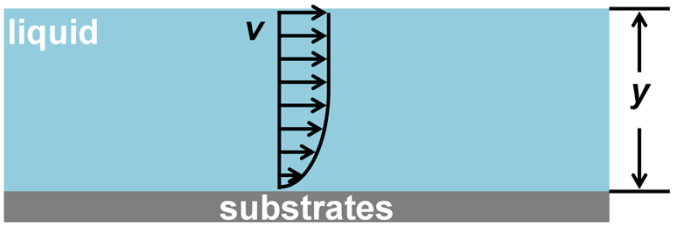

Fig.S3 Illustration of shear stress.

4. PSS/PAH Multilayer and Its AFM Morphology

HG-LbL Process of PSS/PAH Multilayers. The PDDA-modified substrates were rotated at 2400 rpm. The PSS solution (1 mg/mL, pH 9.5) was pumped into the cavity at 30 mL/min for 1 min, and then the electrical machine was raced for 1 min. Deionized water was pumped into the cavity to rinse the substrates for 1 min, and then the electrical machine was kept operating for 1 min. A solution of PAH (1 mg/mL, pH 9.5) was pumped into the cavity at 30 mL/min for 1 min, and then the electrical machine kept rotating for 1 min; the substrates were again rinsed for 1 min. Multilayer assemblies were generated by repeating the four steps above in a cyclic fashion.

LbL dipping assembly process of PSS/PAH Multilayer. The PDDA-modified substrates were immersed in a PSS solution (1 mg/mL, pH 9.5) for 20 min, rinsed with deionized water, followed by dried in a N_2 flow. Then, the substrates were transferred into PAH solution (1 mg/mL, pH 9.5)

for another 20 min, rinsed and dried. Repeat the above steps until the desired bilayer was completed.

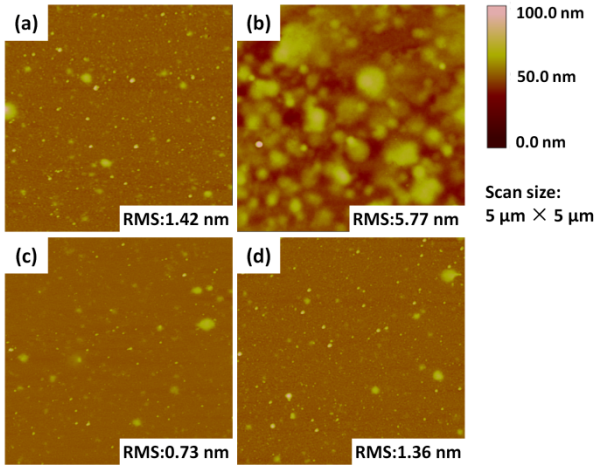

Fig.S4 AFM height images of PSS/PAH–PAA multilayers fabricated by (a)dipping LbL, 4 bilayers; (b) dipping LbL, 7 bilayers; (c)HG-LbL, 4 bilayers and (d)HG-LbL, 7 bilayers.

5. PSS/PAH-PAA Multilayer and Its AFM Morphology

HG-LbL Process of PSS/PAH-PAA Multilayers. The PDDA-modified substrates were rotated at 2400 rpm. The PSS solution (1 mg/mL, pH 8.5) was pumped into the cavity at 30 mL/min for 1 min, and then the electrical machine was raced for 1 min. Deionized water was pumped into the cavity to rinse the substrates for 1 min, and then the electrical machine was raced for 1 min. A solution of PAH-PAA complex (0.93 mg/mL, pH 8.5) was pumped into the cavity at 30 mL/min for 1 min, and then the electrical machine was raced for 1 min; the substrates were again rinsed for 1 min. Multilayer assemblies were generated by repeating the four steps above in a cyclic fashion.

LbL dipping assembly process of PSS/PAH-PAA Multilayer. The PDDA-modified substrates were immersed in a PSS solution (1 mg/mL, pH 8.5) for 20 min, rinsed with deionized water, followed by dried in a N_2 flow. Then, the substrates were transferred into PAH-PAA complex solution (0.93 mg/mL, pH 8.5) for another 20 min, rinsed and dried. Repeat the above steps until the desired bilayer was completed.

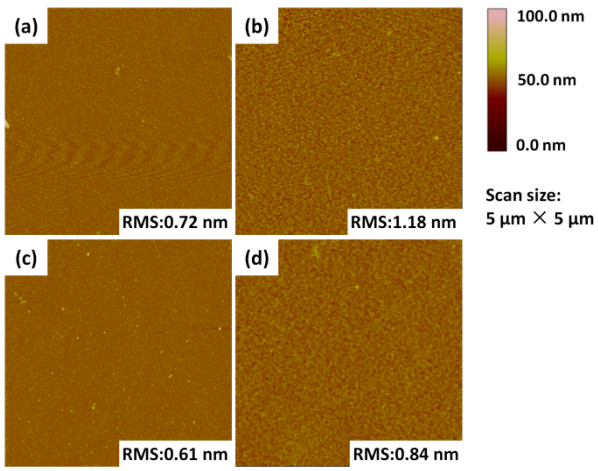

Fig.S5 AFM height images of PSS/PAH multilayers fabricated by (a)dipping LbL, 4 bilayers; (b) dipping LbL, 7 bilayers; (c)HG-LbL, 4 bilayers and (d)HG-LbL, 7 bilayers.

6. Hydrodynamic radius distribution of PAH-Por and PAH/PAA complex.

The characterization of PAH/Por and PAH/PAA by DLS is shown in **Fig. S6** and **Fig. S7**, respectively.DLS was measured at a scattering angle of 90^{o} .

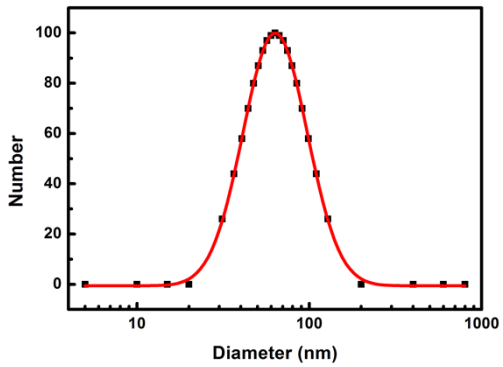

Fig.S6 Hydrodynamic radius distribution curves of PAH-Por complex.

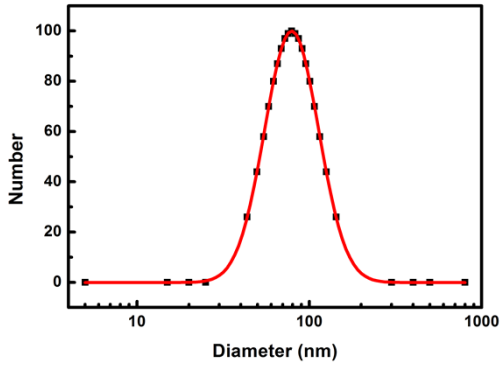

Fig.S7 Hydrodynamic radius distribution curves of PAH-PAA complex.