*Supporting Information*

# Controlled Release from Model Blended Multilayer Films Containing Mixtures of Strong and Weak Polyelectrolytes

*Yeongseon Jang,*[1] *Bulent Akgun,*[2,3] *Hosub Kim,*[1] *Sushil Satija,*[2] *and Kookheon Char*[1,*]

[1]The National Creative Research Initiative Center for Intelligent Hybrids, The WCU Program of Chemical Convergence for Energy & Environment, School of Chemical & Biological Engineering, Seoul National University, Seoul 151-744, Korea

[2]NIST Center for Neutron Research, NIST, Gaithersburg, Maryland 20899-6102, United States

[3]Department of Materials Science and Engineering, University of Maryland, College Park, Maryland 20742, United States

Corresponding Author

*Tel: +82-2-880-7431, fax: +82-2-873-1548, e-mail: khchar@plaza.snu.ac.kr

**Table S1.** Incorporation ratio of PMAA within the blend multilayer films calculated from the film mass obtained from QCM measurements and the IR absorbance peak areas of PMAA chains at 1540 and 1701 $cm^{-1}$.

| **Feed Ratio of PMAA in Anionic Solutions** | **Incorporation Ratio of PMAA in Blend Multilayer Films** |
|---|---|
| 100 | 100 |
| 90 | 91 ± 1.7 |
| 80 | 83.2 ± 3.1 |
| 70 | 73.3 ± 2.6 |

***Calculation on the Amount (wt%) of PMAA incorporated in Blend Multilayer Films:***

The frequency changes ($\Delta f_3/3$) as well as the absorbance peak areas of 100 wt% PMAA layers were measured by QCM and FT-IR. The IR absorbance peak area was calculated by the Gaussian-Lorentz method. Based on the mass and the IR absorbance of pure PMAA layers, the mass and IR absorbance of blend multilayer films were normalized and the incorporated amounts of PMAA in the blend films were calculated.

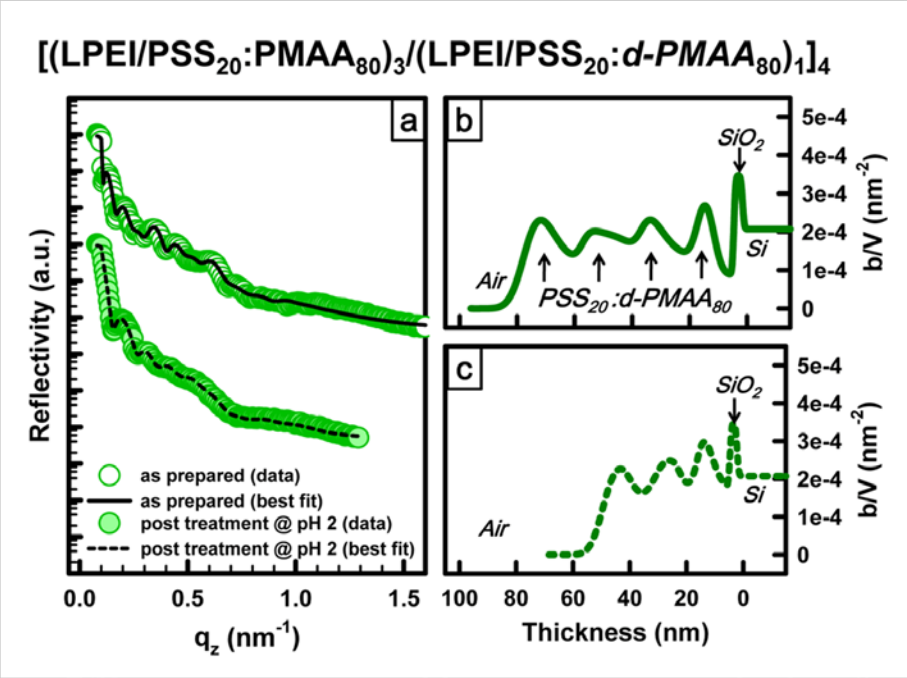


**Figure S1.** Neutron reflectivity (NR) curves with best fits and the SLD profiles of blend multilayer films, $[(LPEI/PSS_{20}:PMAA_{80})_3(LPEI/PSS_{20}:d\text{-}PMAA_{80})_1]_4$. The film initially deposited at pH 5 is represented by open symbols in (a) the reflectivity panels and solid lines in (b) the SLD panels; and the film post-treated at pH 2 for 10 min is represented by closed symbols in (a) the reflectivity panels and dashed lines in (c) SLD panels.

***QCM data reduction***

Every blend multilayer film was deposited on a cleaned Au sensor crystal. Every film was stabilized in pH 5.0 water solution, which is the same pH condition for the film deposition. 0.8 ml of pH 2.0 water was then injected to the sample chamber to initiate the triggered release. As shown in Figure S2, the frequency changes ($\Delta f_n/n$) in all blend multilayer films triggered at pH 2 show the superimposed characteristics with different overtone numbers (n = 3, 5, 7) and dissipation energy less than $2.0\times10^{-6}$. Therefore, the released film masses ($ng\cdot cm^{-2}$) as a function of post-treatment time at pH 2.0 were calculated based on the Sauerbrey equation. Typically, $\Delta f_1$ and $\Delta D_1$ were typically noisy due to insufficient energy trapping. Thus, frequency changes in the third overtone $\Delta f_3/3$ (Hz) were compared with different blend multilayer films. Burst in the release profile of non-blended multilayer film containing LPEI and PMAA only was developed when it triggered at pH 2.0. On the other hand, blended multilayer films containing 10 mol% and 30 mol% PSS have unique release profile followed by instant swelling. In order to normalize the QCM data of each blend multilayer film, the starting point of the release from the films is set to zero based on the absolute frequency stabilized at pH 5.

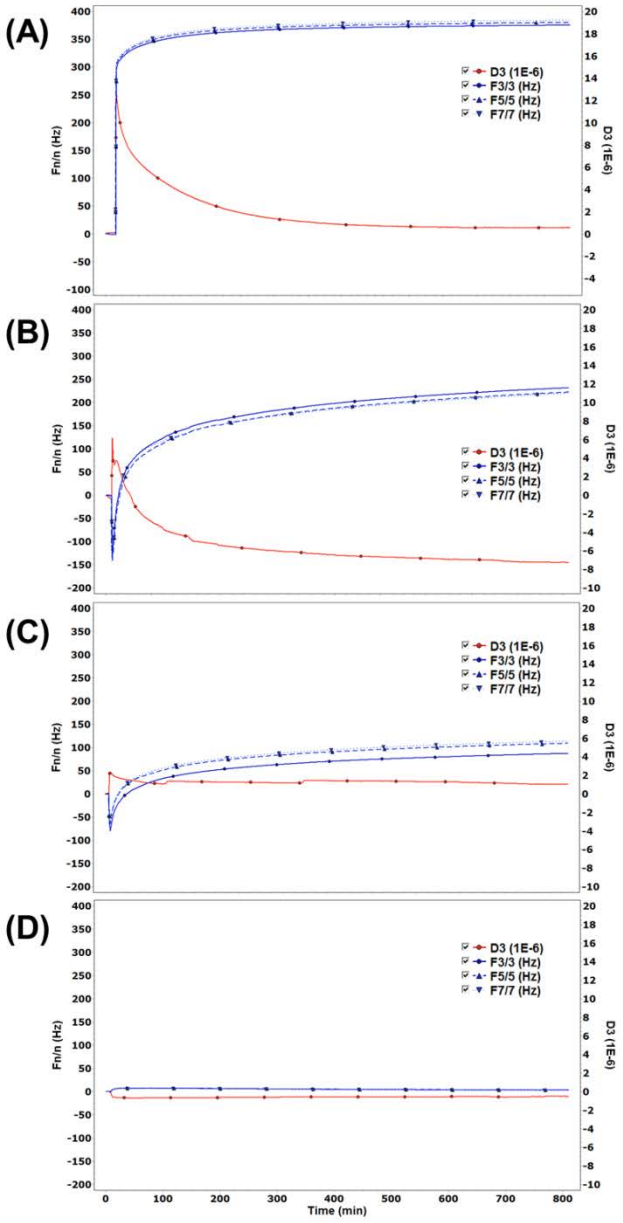


**Figure S2.** QCM raw data of blend multilayer films, $[LPEI/PSS:PMAA]_{16}$ with different blend ratios (PSS:PMAA = (A) 0:100, (B) 10:90, (C) 30:70 and (D) 100:0). Frequency changes (blue) with overtone numbers (n = 3 (circle, solid line), 5 (triangle, dash), 7 (down triangle, dots)) and dissipation changes (red) were monitored as a function of post-treatment time in pH 2.0 water.

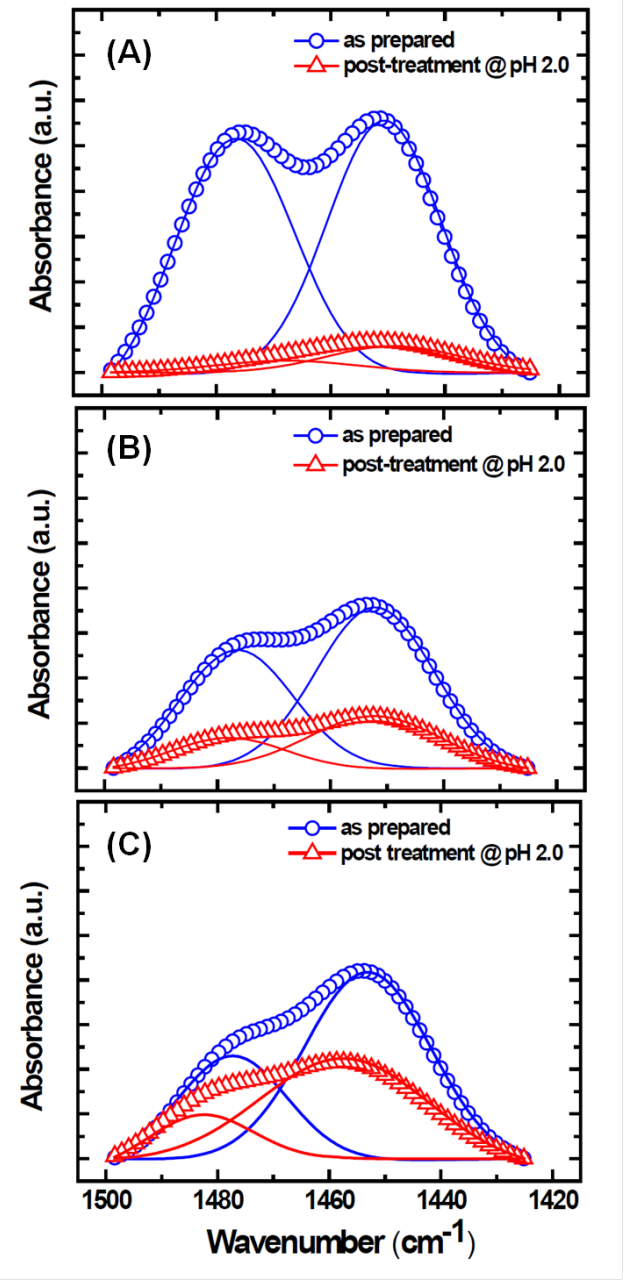


**Figure S3.** FT-IR spectra of LPEI chains, in the regions at 1450 and 1480 $cm^{-1}$ associated with NH bending peak connected to $CH_2$ scissors, for as-prepared films (circle symbols) and the films post-treated at pH 2 (triangle symbols) at different blend ratios in PSS:PMAA polyanion mixtures ((A) 0:100; (B) 10:90; (C) 30:70).