Supplementary Information for

Nanometer-thick lateral polyelectrolyte micropatterns induce macrosopic electro-osmotic chaotic fluid instabilities

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A. Screening of polyelectrolyte combinations

Six different polyelectrolyte combinations (Table S1) were screened, to quickly preselect the best combination for subsequent investigations of the effect of the number of bilayers and the microcontact printing. The criteria for the selection were: higher degree of hydrophobicity of the surface, lower plateau length and the absence of water splitting. A summary of the results are depicted in Figure S2 and Figure S3. On the basis of the screening experiments, the polyelectrolyte pair PAH(pH=10)/PAA(pH=3) was selected for further study.

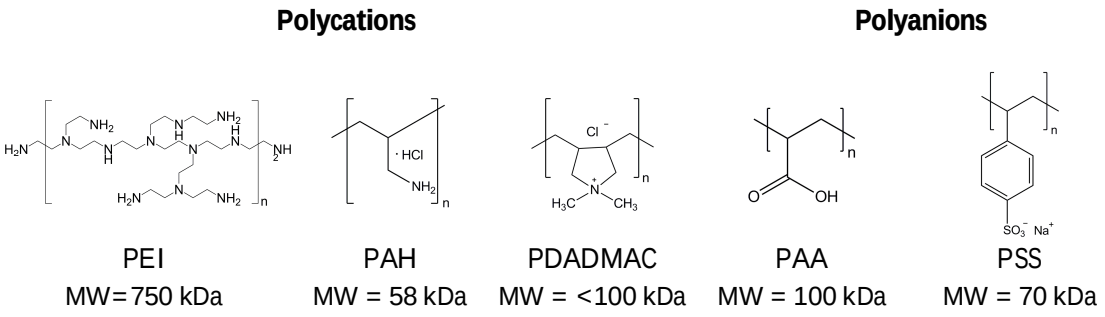

Figure S1. List of polyelectrolytes used for the screening experiments.

Table S1. Combination of polyelectrolytes used for the screening

#	Polycation	Polyanion
1	PAH (pH=10)	PSS
2	PDADMAC	PSS
3	PEI	PSS
4	PAH (pH=10)	PAA (pH=3)
5	PDADMAC	PAA (pH=3)
6	PEI	PAA (pH=3)

(a) (b)

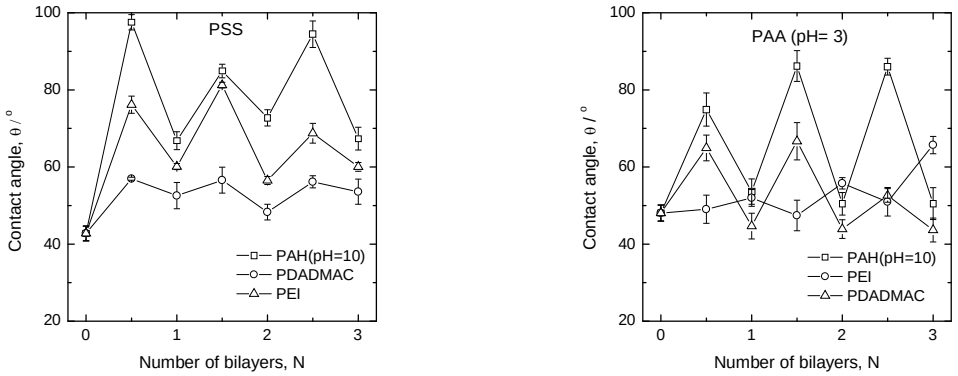

Figure S2. Contact angle results as a function of the number of bilayers of three different polycations in combination with (a) the polyanion PSS, (b) the polyanion PAA (pH =3).

(a) (b)

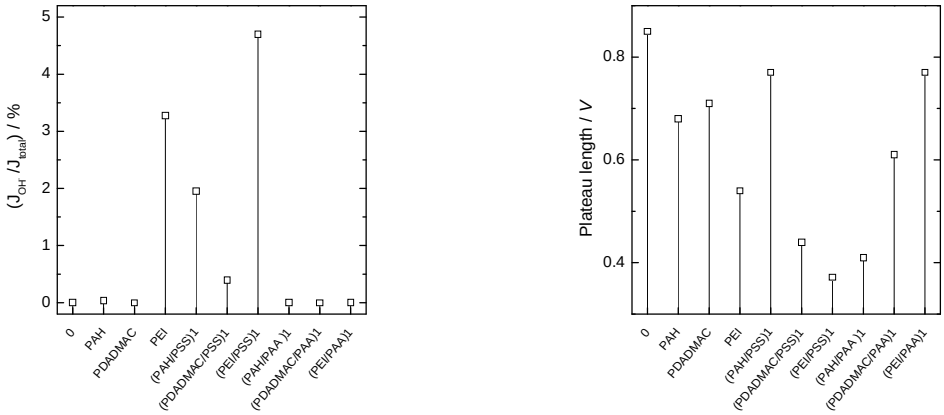

Figure S3. Results obtained from the i-V polarization curves of the screening experiments. (a) Percentage of the current carried by the water splitting products in relation to the total ion flux, during the i-V experiments. (b) Plateau lengths.

B. LBL Surface Analysis (PAH/PAA multilayers on CMX)

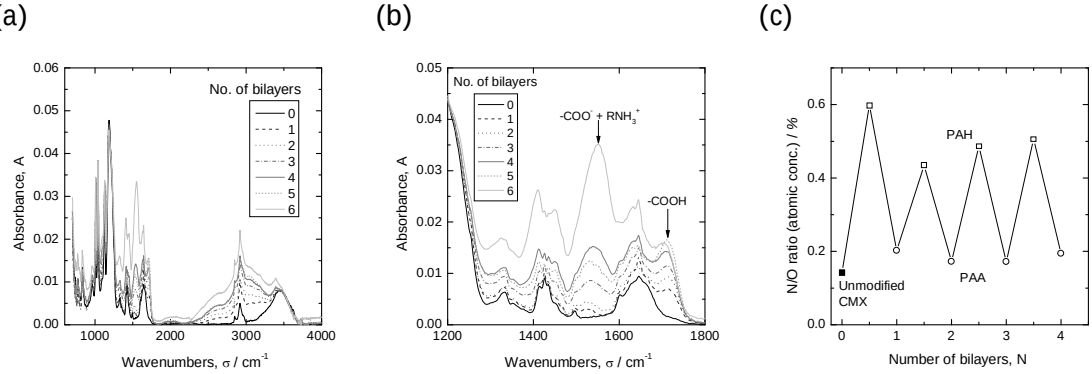

Figure S4. ATR-FTIR spectra of the CMX membrane coated with multilayers of PAH (pH=10) an PAA (pH=3) (a) whole spectra (b) zoomed to wave numbers around 1711 cm^{-1} characteristic for –COOH groups, and around 1551 cm^{-1} which could be an overlay of $-COO^-$ and RNH_3^+ signals. (c) N/O ratio against the number of bilayers obtained from elemental analysis of the XPS results.

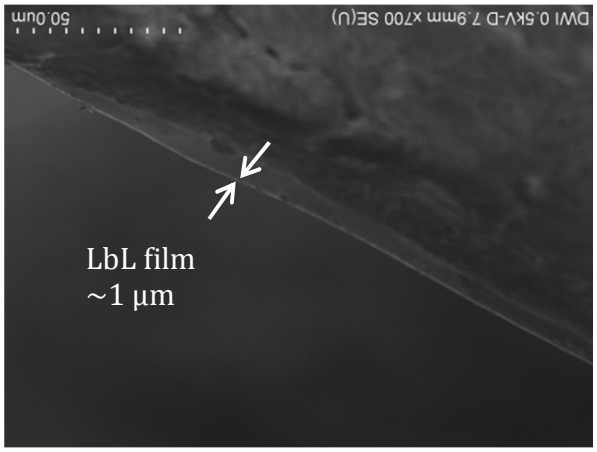

Figure S5. Cross-section scanning electron micrograph of CMX membrane modified with 10 bilayers of PAH (pH = 10) and PAA (pH=3).

C. Microcontact printing

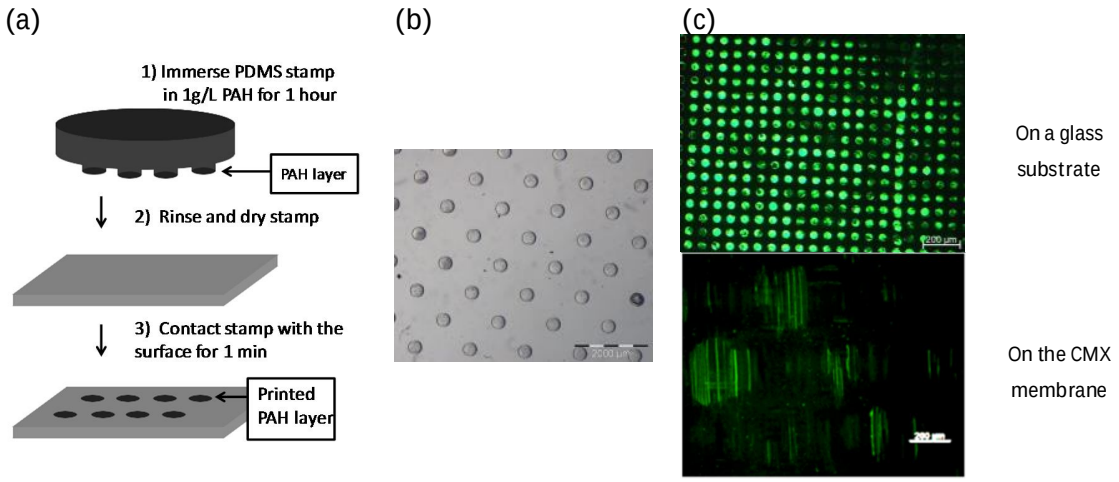

Figure S6. (a) Microcontact printing protocol. (b) A light microscopic image of the PDMS stamp used to print the patterns atop the CMX membrane. (c) Fluorescence micrographs (top) a sample image on a glass substrate; a different PDMS stamp was used to print the patterns (bottom) on CMX membrane.