Supporting Information

Epitope Binning Assay Using an Electron Transfer-Modulated Aptamer Sensor

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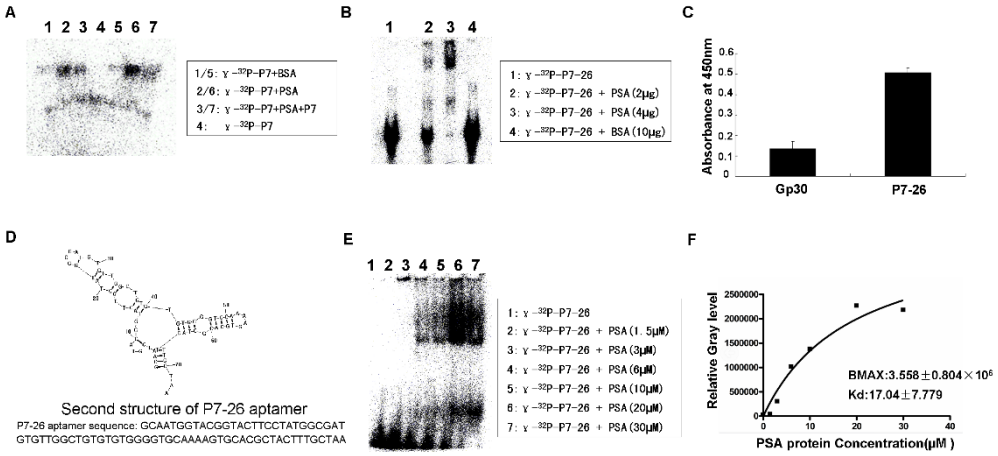
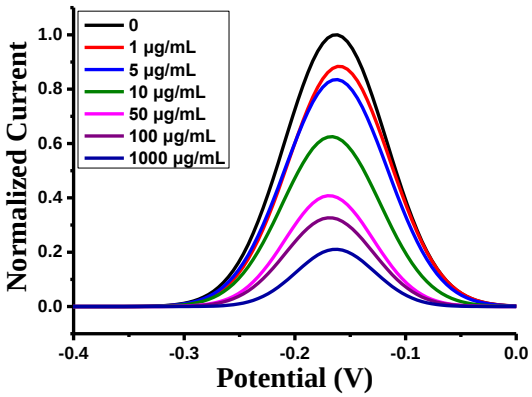
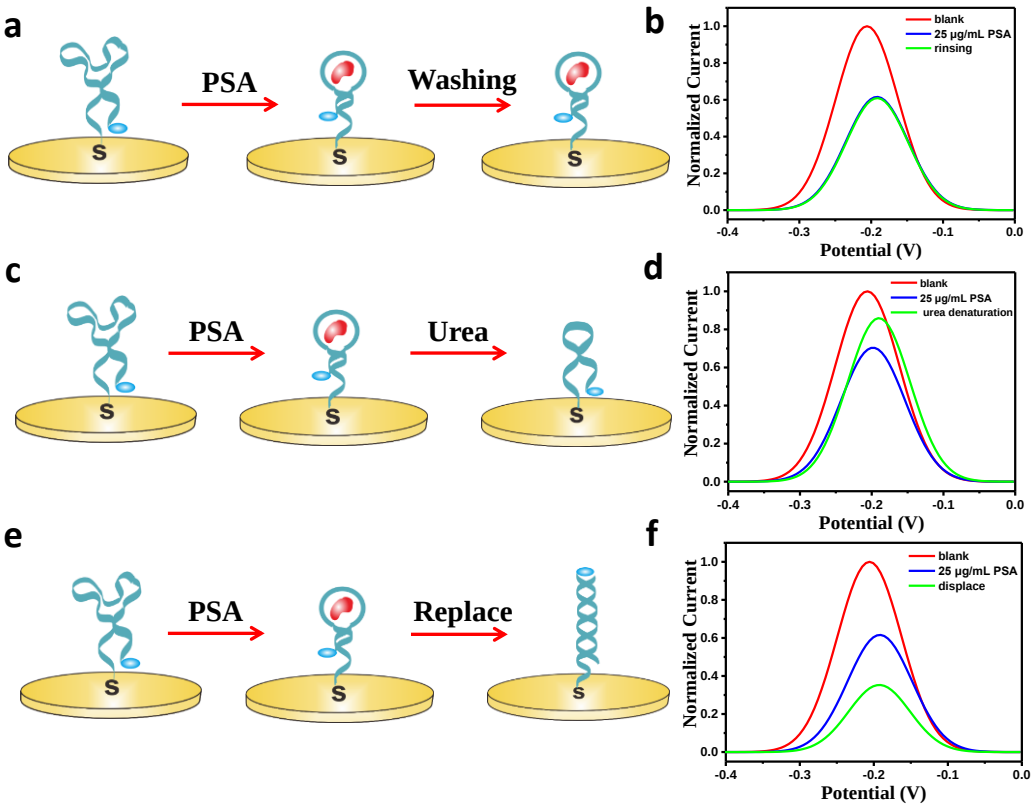


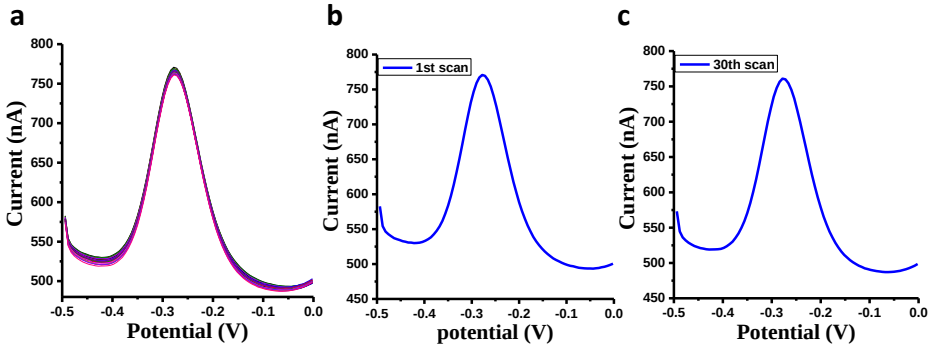
S1 Identification of the binding of the 7th pool and the P7-26 aptamer to PSA. (A) EMSA analysis of the enrichment of the 7th pool. (B) The recognition of the P7-26 aptamer to PSA by EMSA.(C) The binding of the P7-26 aptamer to PSA by ELONA.(D) The primary and secondary structure of P7-26. (E、F) The calculated Kd of the P7-26 aptamer to PSA by EMSA.



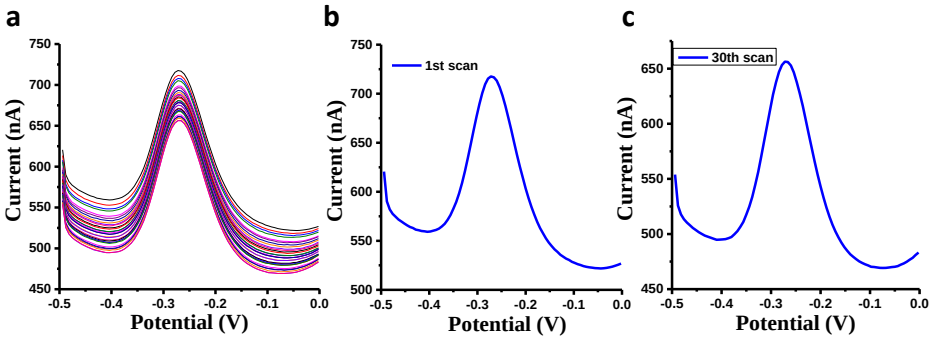
S2. Redox electrochemical signal changes of MB-Apt2 after incubation with PSA of varying concentration.



S3. Verification of the specific binding between Apt2 and PSA. (a) Schematic representation of conformational change of MB-Apt2 before, after PSA binding and followed by Milli-Q water rinsing treatment. (b) Square wave voltammetry of MB-Apt2 before, after PSA binding and followed by Milli-Q water rinsing treatment. (c) Schematic representation of conformational change of MB-Apt2 before, after PSA binding and followed by urea denaturation treatment. (d) Square wave voltammetry of MB-Apt2 before, after PSA binding and followed by urea denaturation treatment. (e)Schematic representation of conformational change of MB-Apt2 before, after PSA binding and followed by complementary DNA replacement. (f) Square wave voltammetry of MB-Apt2 before, after PSA binding and followed by complementary DNA replacement.



S4. The stability of redox electrochemical signal of the unbound MB-Apt2. (a) Square wave voltammetry of MB-Apt2 scanned for 30 cycles. (b-c) The first and 30th scan of MB-Apt2, respectively.



S5. The stability of redox electrochemical signal of the bound MB-Apt2 after PSA binding. (a) Square wave voltammetry of MB-Apt2 scanned for 30 cycles. (b-c) The first and 30th scan of MB-Apt2, respectively.

Supplementary Table S1. Random ssDNA library and primers were used in the experiments

Name	Sequence
GP30	5′ -G CAATGGTACGGTACTTCC-N30-CAAAAGTGCACGCTACTTTGCTAA-3′
Plong-1	5′ - GCAATGGTACGGTACTTCC-3′
P11	5′ - TTAGCAAAGTAGCGTGCACTTTTG-3′
Pstem-loop	5′ -GCTAAGCGGGTGGGACTTCCTAGTCCCACCCGCTTAGCAAAGTAGCGTGCACTTTTG-3′

Supplementary Table S2. Six candidate sequences

Aptamer	Sequence
P7-7	5′ -GCAATGGTACGGTACTTCC**TCTTGGGGTCTTATGTTCGTTCACGGTTGC**CAAAAGTGCACGCTACTTTGCTAA-3′
P7-10	5′ -GCAATGGTACGGTACTTCC**CCGGTGCTTATTCCTGTCTCCTCTCTGCGT**CAAAAGTGCACGCTACTTTGCTAA-3′
P7-19	5′ -GCAATGGTACGGTACTTCC**TGTGTGCTGGGATTCAGTGTTGTTGTTTGTC**CAAAAGTGCACGCTACTTTGCTAA-3′
P7-22	5′ -GCAATGGTACGGTACTTCC**TTATTGTATGTCAGTTGGATGTATGGGTAG**CAAAAGTGCACGCTACTTTGCTAA -3′
P7-26	5′ -GCAATGGTACGGTACTTCC**TATGGCGATGTGTTGGCTGTGTGTGGGGTG**CAAAAGTGCACGCTACTTTGCTAA-3′
P7-30	5′ -GCAATGGTACGGTACTTCC**TGGTGTTGTTAGCGTTGTTTTCTTGTGTTTG**CAAAAGTGCACGCTACTTTAATCG-3′

The random rigion of the aptamers were shown in bold with underline