

Electronic Supplementary Information

**Magnetic separation-assisted cluster-amplified versatile fluorescent aptasensors for sensitive detection of target biomolecules**

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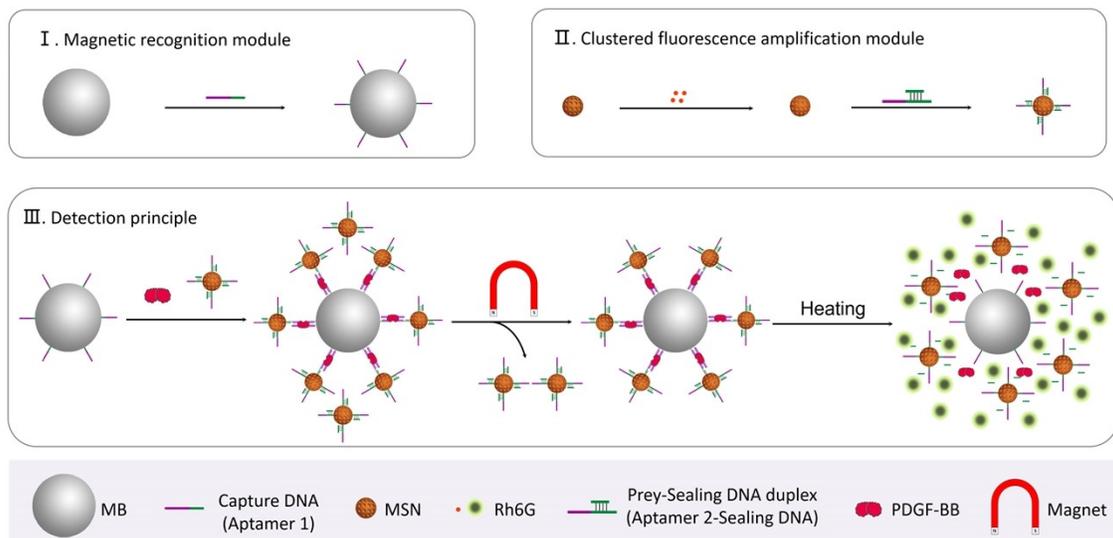
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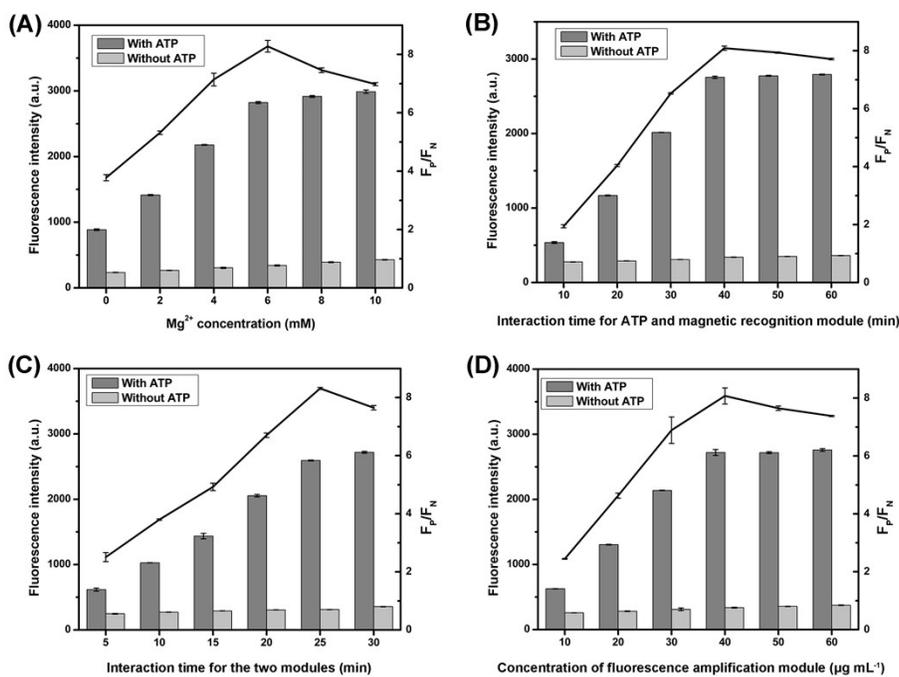
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**Synthesis of aminated MSNs.** Co-condensation method was adopted to synthesize the aminated MSNs (MSN-NH<sub>2</sub>). Aqueous sodium hydroxide (1.5 M, 2.4 mL) was added to 240 mL of Cetyltrimethylammonium bromide (CTAB) solution (5.7 mM) and stirred for 5 min. Then heated the mixture to 80 °C and maintained for 30 min. Subsequently, 2.4 mL of Tetraethoxysilane (TEOS) and 0.5 mL of (3-Aminopropyl)-triethoxysilane (APTES) were dropwise added sequentially. Then stabilized the mixture at 80 °C for 2 h and cooled it to room temperature. Separated the resultant precipitate from the mixture by centrifugation, and washed it using ethanol and water. The internal CTAB was removed by refluxing it in a mixed liquor of methanol (80 mL) and hydrochloric acid solution (37.4 %, 1.0 mL) for 15 h. Finally, rinsed the MSN-NH<sub>2</sub> by ethanol and dried it for storage.

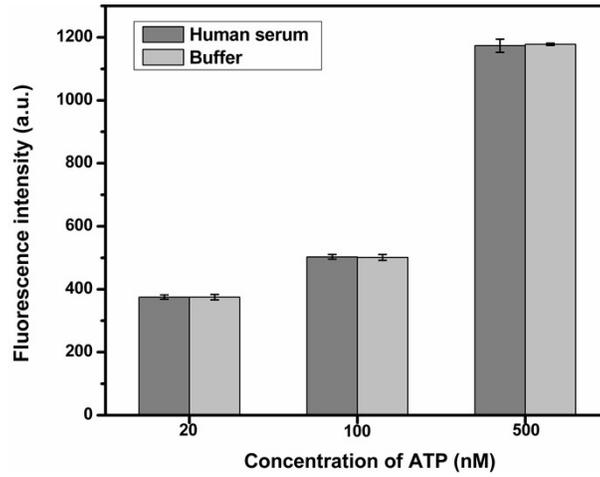
## Supplementary figures



**Fig. S1.** Schematic illustration of the modular aptasensor for PDGF-BB detection.



**Fig. S2.** Effect of (A)  $Mg^{2+}$  concentration, (B) interaction time for ATP and magnetic recognition module, (C) interaction time for fluorescence amplification module and magnetic recognition module, (D) concentration of fluorescence amplification module on the fluorescence intensity and  $F/F_0$ . Error bars showed the standard deviation of three parallel experiments.



**Fig. S3.** The fluorescence intensities for different concentrations of ATP in reaction buffer and human serum. (Error bars showed the standard deviations of three independent experiments.)

## Supplementary tables

Name	Sequence (5'→3')
Aptamer	GAGAGAACCTGGGGGAGTATTGCGGAGGAAGGT
Capture DNA	CCCAGGTTCTCTCTCACACATCTGT-biotin
Prey DNA	GAGAGAACCTGGGGATATGCTAGACTATCG-NH <sub>2</sub>
Sealing DNA	CGATAGTCTAGCATA

**Table S1** Sequences of oligonucleotides used for ATP detection.

True concentration (nM)	Intra-day measurement (nM)	RSD (n = 3)	Inter-day measurement (nM)	RSD (n = 3)
20	20.3	3.1 %	19.2	5.5 %
	21.5		21.2	
	20.5		19.4	
100	104	3.1 %	103	4.6 %
	105		103	
	99		95	
500	508	1.0 %	488	2.9 %
	498		516	
	506		508	

**Table S2** Intra-day and Inter-day detection for ATP of different concentrations.

True concentration (nM)	F - F <sub>0</sub> (a.u.)	Calculated concentration (nM)	Average recovery	RSD (n = 3)
20	37.87	19.0	98 %	4.0 %
	40.27	20.5		
	38.51	19.4		
100	174.1	104	102 %	3.2 %
	172.5	103		
	164.5	98		
500	821.7	508	101 %	2.1 %
	826.5	511		
	794.5	491		

**Table S3** Recoveries of ATP in 10 % human serum.

Name	Sequence (5'→3')
Aptamer	GCTCGGAGTCCGTGGTAGGGCAGGTTGGGGTGACT
Capture DNA	ACGGACTCCGAGCTCACACATCTGT-biotin
Prey DNA	GCTCGGAGTCCGTGATATGCTAGACTATCG-NH <sub>2</sub>
Sealing DNA	CGATAGTCTAGCATA

**Table S4** Sequences of oligonucleotides used for thrombin detection.

True concentration (nM)	F - F <sub>0</sub> (a.u.)	Calculated concentration (nM)	Average recovery	RSD (n = 3)
	10.37	0.036		
0.04	11.39	0.038	96 %	6.6 %
	12.92	0.041		
	195.7	0.4		
0.4	180.4	0.37	95 %	4.6 %
	180.4	0.37		
	2079	4.1		
4.0	1927	3.8	98 %	3.9 %
	1978	3.9		

**Table S5** Detection of thrombin of different concentrations in 10 % human serum.

Name	Sequence (5'→3')
Aptamer 1	CAGGCTACGGCACGTAGAGCATCACCATGATCCTGTCACACATCTGT-biotin
Aptamer 2	CAGGCTACGGCACGTAGAGCATCACCATGATCCTGGATCTACTAGACTATCG-NH <sub>2</sub>
Sealing DNA	CGATAGTCTAGCATA

**Table S6** Sequences of oligonucleotides used for PDGF-BB detection.

True concentration (nM)	F - F <sub>0</sub> (a.u.)	Calculated concentration (nM)	Average recovery	RSD (n = 3)
0.02	10.89	0.019	95 %	5.2 %
	10.57	0.018		
	11.22	0.020		
0.2	68.89	0.20	102 %	2.8 %
	68.89	0.20		
	72.09	0.21		
2.0	677.6	2.1	98 %	5.8 %
	613.6	1.9		
	613.6	1.9		

**Table S7** Detection of PDGF-BB of different concentrations in 10 % human serum.