**Goal:** To develop biomolecular and biomimetic materials whose multiple functions can be programmed in diverse ways.

From affinity aptamers

To intelligent nanobiomaterials

To tissue-like biomaterials
On-Demand Release of Multiple Protein Drugs For Tissue Engineering and Regenerative Medicine

J. R. Soc. Interface (2011) 8, 153–170
**Principle:** Molecular Recognition


Nucleic Acid Aptamers

Single-stranded oligonucleotides screened from the library of synthetic oligonucleotides

Nucleic Acid Aptamers

- High specificity
- High affinity
- Little immunogenicity
- Small size
- Easy synthesis
- Tolerant of harsh chemical/physical conditions
  - High resistance against nuclease degradation
  - Controllable reversibility in molecular recognition

Synthesis of Aptamer-Functionalized Hydrogels via Free Radical Polymerization

Effect of Binding Affinity
on Retention/Release of Growth Factors

\[ \lambda V_g \frac{dP^{(s)}}{dt} = hS \left[ P^{(g)} - P^{(s)} - P_{\infty}^{(g)} + P_{\infty}^{(s)} \right], \]

\[ \frac{dP^{(g)}}{dt} = -kAP^{(g)} + k'C - \lambda \frac{dP^{(s)}}{dt}, \]

\[ \frac{dA}{dt} = -kAP^{(g)} + k'C, \]

\[ \frac{dC}{dt} = kAP^{(g)} - k'C, \]

Pros & Cons

**Good:**
- Aptamers were able to retain growth factors in the hydrogels;
- The retention/release could be modulated by varying the binding affinity of the aptamer.

**Bad:**
- Some growth factors were significantly or completely denatured during the synthesis of hydrogels.
- We also tried other methods like photoinitiated polymerization. It did not work well for us.
Synthesis of Aptamer-Functionalized Hydrogels Using Thermoresponsive Solutions

Release of Growth Factors from Aptamer-Functionalized Agarose Hydrogels

Retention

Programmable Release


Region for Hybridization

A

B

C

D
Length of Hybridization

Apt-1: 5'- ACAGGCTACGGCAGTACGATCCATCATGATCCTG -3'
CO-4: 3' - GTAGTGACTAGGAC -5'
CO-5: 3' - CGTGCATCTCTGAGTCGATTAGGAC -5'
CO-6: 3' - TGTCCGATGCCATCTCAGTGACTAGGAC -5'

![Graph showing association and dissociation of response over time.](image-url)
Synergistic Effect: Tail + Length

Apt-4:
5'- ACAGGCTACGGCACGCAGACATCACCATGATCCTGTGACTTGAGCAAAAT -3’
3’- ACTGAACCTCGTTTTA -5’
3’-ACACTGAACCTCGTTTTA -5’
3’-GGACACTGAACCTCGTTTTA -5’
3’- AGTGGTACTAGGACACTGAACTCGTTTTA -5’
3’- TGCATCTCGTAGTGGTACTAGGACACTGAACTCGTTTTA -5’
3’- TCCGATGCGCGATCCTCGTAGTGGTACTAGGACACTGAACTCGTTTTA -5’
SPR Analysis of Programmable Molecular Recognition

![Graph showing association and dissociation of aptamers with and without a trigger.](image)
Programmable Release of Multiple Protein Drugs

Two different protein drugs were programmed to release at days 5 and 10.

Structures of Superporous Hydrogels

- Nonporous
- w/o aptamer
- w/ aptamer

Histogram of Pore size [µm]:
- 0-9%
- 10-19%
- 20-29%
- 30-39%
- 40-49%
- 50-59%
- 60-69%
- 70-79%
- 80-89%
- 90-99%
Hydrogels for the Delivery of Protein Drugs - Past & Current

Past

*Nature; 1976 (263): 797-800*

Current Biomaterials 2014; 35(27), 8040-8048

Experimental & Molecular Medicine 2012 (44), 350-355
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