Ion-controlled three-color fluorescent telomeric switch on DNA origami

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DNA origami nanostructures are excellent substrates to arrange and analyze different molecules with a high local control. [1] Förster Resonance Energy Transfer (FRET) is an important process which can be used to study conformational changes of biomolecules. We analyzed the conformational changes of telomeric DNA from a random coil into guanine (G) quadruplexes in presence of monovalent cations using FRET. We observed that in contrast to the free telomere in solution, the G-quadruplex formation becomes selective towards K^+ when the telomeric DNA is attached to DNA origami structures. Sensitive K^+ sensing can be achieved even in solutions containing Na^+ at a concentration as high as 145 mM, which represents the physiologically relevant concentration. [2] To use this system as a biosensor, the reversibility of the G-quadruplex formation was extensively studied using fluorescence spectroscopy. The G-quadruplex unfolds and returns to its initial conformation when K^+ is removed. This is feasible with an ion-complexing agent. After a subsequent K^+ addition the G-quadruplex folds again. This folding and unfolding cycle can be repeated several times (Figure 1A). Furthermore, we designed a switchable photonic-wire like system on DNA origami structures by attaching a third dye to the two-color FRET system (Figure 1B). [3]

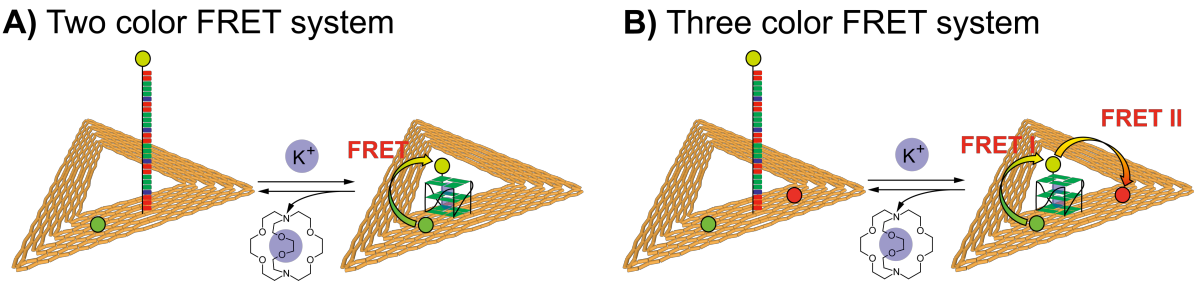

Fig. 1: Schematic illustration of studied systems based on FRET measurements. A) Two-color FRET system with telomeric DNA as an in-situ switch. Fluorescein (green circle) was used as the donor molecule and Cyanine3 (yellow circle) as the acceptor molecule. B) Switchable three color-FRET cascade on DNA origami structures with Fluorescein (green circle) as donor, Cyanine3 (yellow circle) as transmitter and Cyanine5 (red circle) as acceptor molecules.

[1] P. Rothemund Nature 440 (2006) 297-302 and I. Bald, A. Keller Molecules 19(9) (2014) 13803-13823.
[2] L. Olejko, P. J. Cywinski I. Bald, Angew. Chem. Int. Ed. 54(2) (2015) 673-677.
[3] L. Olejko, P. J. Cywinski I. Bald, submitted.