

Nanomaterial Mediated Luminescent Non-canonical DNA Structures

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DNA has been employed as a versatile building block for assembling nanomaterials due to its distinct programmability and predictability. With its highly predictable base-pairings, low cost, ease of synthesis, and biocompatibility, DNA has become the scaffold of choice to assemble nanomaterials for ever-increasing applications in sensing, imaging, and drug delivery, to name a few. However, we have identified that immobilized nanomaterials can function as nano rivets linking DNA scaffolds. One such fluorescent nanomaterial is DNA-stabilized silver nanoclusters (DNA/AgNCs). DNA/AgNCs are <2nm in size and have impressive fluorescent properties. Our work with this promising DNA nanomaterial has established principles to rationally design DNA secondary structures for synthesizing silver nanoclusters (AgNCs) with predictable fluorescent properties. We have defined the arrangement of AgNCs within the DNA template and how the AgNCs influence the secondary structure of the DNAs. Here, we identified a non-canonical supramolecular DNA structure driven by the luminescent AgNCs through metal-mediated base pairing. Through detailed investigations, we identified AgNCs-mediated cytosine-Ag-cytosine bridging between two six-cytosine loop-bearing hairpin DNAs.¹ Based on the in-depth understanding of the mutual influence between the host DNA and the associated nanomaterial, AgNCs, on the properties of each other, we have developed a novel strategy for designing DNA nanosensors that can be reversibly turned on/off in response to pH or change fluorescence color when sensing reactive oxygen species (ROS) over multiple cycles.²

Given the unique aptamer properties of DNA, which allow it to bind specifically to a vast array of molecules with very high specificity, we aim to extend our research in DNA/AgNCs for environmental monitoring. The specificity of DNA aptamers can be harnessed to detect pollutants, antibiotics, heavy metals, toxins, and other environmental hazards with high sensitivity and selectivity. By integrating the fluorescent capabilities of DNA/AgNCs with the binding specificity of DNA aptamers, we envision the development of nanosensors capable of detecting and quantifying environmental contaminants.

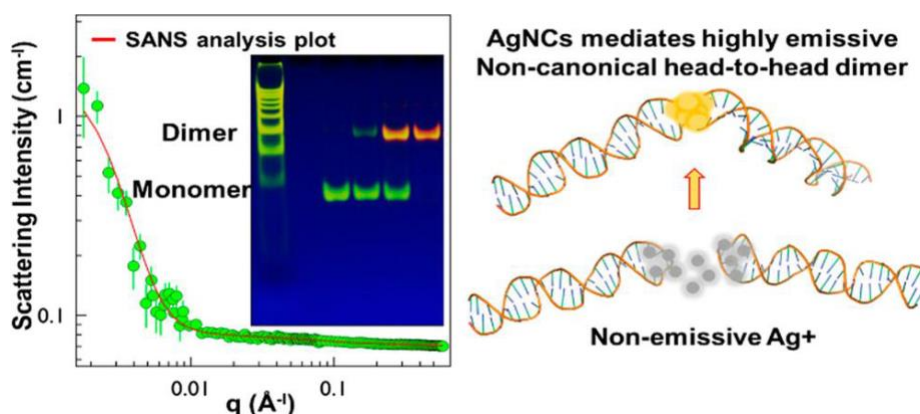


Figure 1: Silver Nanoclusters mediated assembly of supramolecular DNA structure.

1. Shah, P., et.al., Noncanonical Head-to-Head Hairpin DNA Dimerization Is Essential for the Synthesis of Orange Emissive Silver Nanoclusters, *ACS Nano*, 2020, **14 (7)**, 8697-8706.
2. Nagda, R., et.al., Silver Nanoclusters Serve as Fluorescent Rivets Linking Hoogsteen Triplex DNA and Hairpin-Loop DNA Structures. *ACS Nano*, 2022, **16(8)**, 13211-13222.
3. H. C. Yadavalli, et. al, Energy Transfer Between i-Motif DNA Encapsulated Silver Nanoclusters and Fluorescein Amidite Efficiently Visualizes the Redox State of Live Cells. *Small* 2024, 2401629.