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Light-emitting crystals of aptamer-hybrid organic semiconductor signaling on human cells expressing EpCAM

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ABSTRACT

In this study, we successfully performed protein-cell recognition by combining an organic light-emitting diode (OLED) material and an aptamer with a three-dimensional DNA structure. Tris (8-hydroxyquinoline) aluminum (Alq_3), one of the most widely used green OLED components used in displays, was chosen as an emitter. Epithelial cell adhesion molecule (EpCAM) was selected as a target because it is well-established in cancer studies. The generated micrometer-sized particles exhibited increased luminous intensity based on the specific recognition to proteins and cells on hybrid interfaces, proving that the aptamer maintained its three-dimensional structure while binding to its target. Interestingly, only human cells expressing EpCAMs were distinguishable, whereas mouse carcinoma cells were not. Furthermore, use of graphene oxide as a quencher made these aptamer-Alq_3 microrods 2-fold more effective at enhancing the fluorescence signal upon interaction with human oral squamous carcinoma cells. This new approach, which imparts biometric functionality to organic semiconductor materials, will provide a new platform for the evolution of OLED hybrid materials.

Introduction

The recent rapid developments in smart display devices have enabled the production of promising display materials for electronic and optoelectronic research.[1–4] Among them, the organic light-emitting diode (OLED) was first developed using tris(8-hydroxyquinoline) aluminum (Alq_3) in 1987[5], and has since then become a key material in the display industry. It has the advantages of having an excellent contrast ratio, degree of bending, lightweight, and thinness.[6–8] Attempts have been made to import thin biometric films into multilayered-display devices, often referred to as BioLED.[9–11] Recently, DNA oligomers were embedded into an OLED material, Alq_3, to enable DNA-DNA recognition and chemical-DNA interaction.[10–14] DNA recognition triggered photoluminescence enhancement, as reflected in enhanced inhibition of the non-radiative dissipation of Alq_3 molecules,[10] and specific mercury ion-DNA interaction, on the other hand, resulted in decreased luminescence.[12] By leveraging the waveguide effect of one-dimensional DNA-hybrid Alq_3 crystals, the remote identification of DNA-DNA recognition also became possible.[13] Attempts to fuse DNA with OLED materials were successful. However, in order to expand OLED materials to other fields such as medicine or biotechnology, investigation into the applications at the protein and cell level has been much desired.

We first adopted aptamers as bio-recognition modules for OLED hybrid materials. Aptamers are ssDNAs/RNAs with long nucleic acid sequences that exhibit three-dimensional structures and bind specifically to target proteins.[15–19] These aptamers have been widely used in medicine, biotechnology, and biosensors because of their high stability, binding affinity, selectivity, and the ability for chemical modification.[20–24] Hence, aptamers have been combined with Alq_3 forming hybrid interfaces on their surfaces to function as bio-recognitive microparticles with cellular-scale dimensions.

In this study, we selected EpCAM, a protein that is well established in aptamer-protein studies[25–27], as a target of aptamer.

Abbreviations: OLED, organic light-emitting diode; Alq_3, Tris (8-hydroxyquinoline) aluminum; EpCAM, epithelial cell adhesion molecule; Cy3, cyanine3; PL, photoluminescence; CTAB, cetyltrimethylammonium bromide; SDS, sodium dodecyl sulfate; CLSM, confocal laser scanning microscopy; EGFR, human epithelial growth factor; IgG, anti-human immunoglobulin G; HSC-3, human oral squamous carcinoma cells; CMT-93, mouse rectal carcinoma cell line; OMECs, Human oral mucosal epithelial cells.

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