## Triple-Peak Photoluminescence of DNA-Hybrid Alq3 Crystals Emitting a Depressed Single Peak upon Bio-Recognition

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**ABSTRACT:** The green organic semiconductor, tris-(8-hydroxyquinoline)aluminum (Alq<sub>3</sub>), was hybridized with DNA growing in the shape of hexagonal prismatic crystals. In this study, we applied hydrodynamic flow to the fabrication of Alq<sub>3</sub> crystals doped with DNA molecules. The hydrodynamic flow in the Taylor–Couette reactor induced nanoscale pores in the Alq<sub>3</sub> crystals, especially at the side part of the particles. The particles exhibited distinctly different photoluminescence emissions divided into three parts compared to common Alq<sub>3</sub>-DNA hybrid crystals. We named this particle a "three-photonic-unit". After treatment with complementary target DNA, the three-photonic-unit Alq<sub>3</sub> particles doped with DNAs were found to emit depressed luminescence from side parts of the particles. This novel phenomenon would expand the technological value of these hybrid crystals with divided photoluminescence emissions toward a wider range of bio-photonic applications.

**KEYWORDS:** three-photonic-unit Alq<sub>3</sub> particles, hydrodynamic flow, Taylor–Couette reactor, photoluminescence emission, treatment with perfect-matched target DNA

## 1. INTRODUCTION

Self-assembly of the  $\pi$ -conjugated organic molecules was a promising fabrication technique in medicine,<sup>1</sup> catalysis,<sup>2</sup> energy conversion,<sup>3</sup> and sensing applications.<sup>4</sup> Structures with various shapes and sizes had been assembled for unique functional devices. Assembly on the liquid–liquid interface was a versatile method to fabricate functional materials.<sup>5</sup> Assembly was affected by various external factors such as electric fields,<sup>6</sup> magnetic fields,<sup>7</sup> and ultrasound.<sup>8</sup> Liquid–liquid interfacial assemblies had been attempted with respect to reaction conditions including organic/aqueous phase ratio,<sup>9</sup> temperature,<sup>10</sup> solvent, and solubility of precursors to construct functional materials. Such various factors were applied to research strategies for enhancing the functionalities of organic self-assembly materials.

Circular Couette flow had been studied since Taylor's experiments and analysis in 1923.<sup>11–14</sup> Most studies had focused on the stability of the flow. Little attention has been paid to the velocity field generated in the annulus.<sup>15</sup> However, an understanding of the velocity field was important to engineering applications of the flow. It was developed to control the growth rate and size of crystals using flow<sup>16</sup> and reactors with high efficiency of products. For example, Bose–

Einstein condensation manipulated biomolecules, colloids, and cells and enhanced crystallinity of polymers.

One of the organic light-emitting diode (OLED) materials, tris-(8-hydroxyquinoline)aluminum (Alq<sub>3</sub>) has the characteristic of growing in the shape of hexagonal prismatic crystals.<sup>17</sup> Since the report of the growth of long hexagonal rods by the microemulsion method,<sup>18</sup> the hybridization of DNA-doped organic semiconductors was recently reported for the first time.<sup>19</sup> Hybridized organic material showed novel biorecognition properties that the specific DNA–DNA recognition triggers photoluminescence (PL) enhancement.<sup>20</sup> Several studies were reported to utilize the physical and chemical properties of DNA, the biological information carrier, as a molecule with various functions.<sup>21–23</sup> The DNA molecule was widely combined with  $\pi$ -conjugated organic semiconductors and served as an efficient receptor component for chemical/biological target recognition.<sup>24–26</sup> The behavior

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