NANO LETTERS

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Polarized Polymer Rectangles Featuring Long-Range Ordered π-Conjugation for Anisotropic Responses to Light and Tensile Vectors

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Cite This: https://doi.org/10.1021/acs.nanolett.5c01422 Read Online

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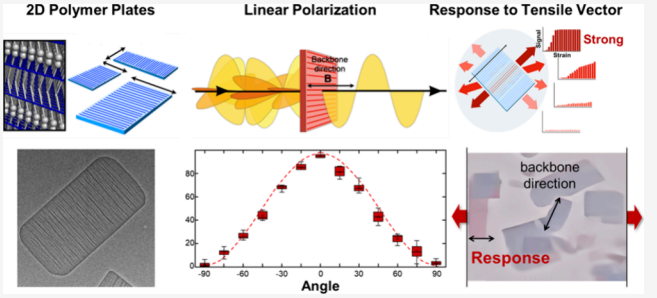
ABSTRACT: Micrometer-scale polarized polymeric rectangles with nanometer-scale thicknesses were fabricated through the simple reprecipitation of selected diacetylenic surfactants, exhibiting anisotropic responses to directional external fields. These 2D rectangular plates feature linearly conjugated poly(diacetylene) backbones aligned along their shorter side over large domains, enabling unique angle-dependent polarization of visible light and fluorescent emission. The conformity to Malus's law confirms the reliability of the linear polarization demonstrated by these plates. In addition to optical anisotropy, these polymer rectangles exhibit an orientation-dependent mechanical response. When tensile force is applied parallel to the shorter side or polymer backbone, the plates undergo a distinct visual color transition; however, little to no change occurs when the force is applied perpendicularly. Such an anisotropic response behavior of 2D rectangular plates is further validated by molecular dynamics simulations. This work provides a strategic framework for materials chemistry design that enables optical reflection of orientation-dependent external fields.



KEYWORDS: *microrectangular 2D plates, nanometer thickness, linear π-conjugation, light polarization, tensile vectors, orientation-dependent signals*

π-Conjugated polymers have gained significant attention as a class of versatile materials in organic electronics, biomedical imaging, and treatment, as well as sensing technologies. These polymeric materials have been widely recognized for their π-conjugated backbones with delocalized electronic structures which impart various optoelectronic properties.[1−4] The conjugated polymers are highly sensitive to external stimuli including physical, chemical, and biological inputs, resulting in specific changes in optoelectronic signals.[5−8] In order to effectively capture field-dependent response characteristics, these polymers need to reflect both the magnitude and orientation of the stimuli satisfying activation thresholds.[9,10] Precise controls of intermolecular interaction, molecular arrangement, and assembled structure in the conjugated polymers are much desired to meet functional performances under demanding constituents.[11] Thereby, the delicate tailoring of responsive conjugated polymers is crucial for the development of field-specific functionality.

Polydiacetylenes (PDAs) are an interesting class of conjugated polymers due to their pronounced colorimetric transitions from blue to red under external stimuli.[12−17] Unique π-conjugated linear backbones readily formed via topochemical photopolymerization undergo changes in the delocalized electronic structure against stimuli imposed, which is characterized by shifts in the HOMO−LUMO energy level detectable both visually and spectroscopically.[18−26] PDAs have been studied for their technical advantages including controlled morphologies, such as Langmuir−Blodgett film, vesicles, wire, and other structures,[27−32] and molecular receptor designs for specific and quantitative detections. Yet, architecture exhibiting colorimetric signals reflecting vectorial stimuli has not been developed.

Recently, various progresses have been reported based on integrated chips for the purpose of the development of sensor devices for mechanical response vectors. However, these devices demand not only complex fabrication in the micro- and nanoscales but also intricate packaging procedures.[33−38] There exists a stark need for a much simplified process to achieve vectorial responsivity.

In this research, we report the fabrication of micrometer-scale rectangular plates (2D plates) of PDAs with nanometer-scale thickness through a reprecipitation process using selected

Received: March 4, 2025
Revised: March 20, 2025
Accepted: March 20, 2025

https://doi.org/10.1021/acs.nanolett.5c01422
Nano Lett. XXXX, XXX, XXX−XXX