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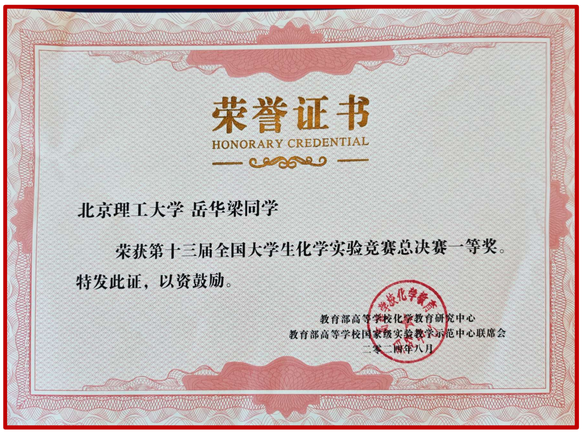

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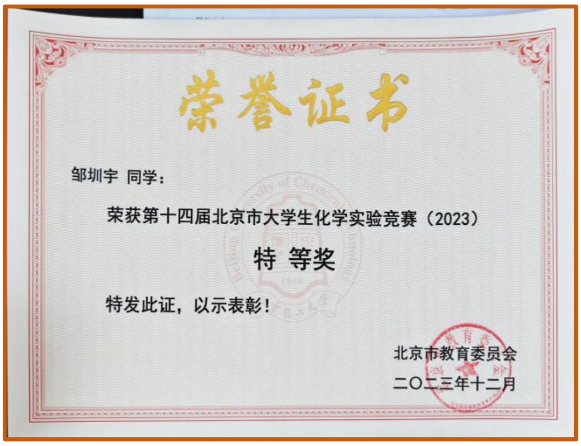

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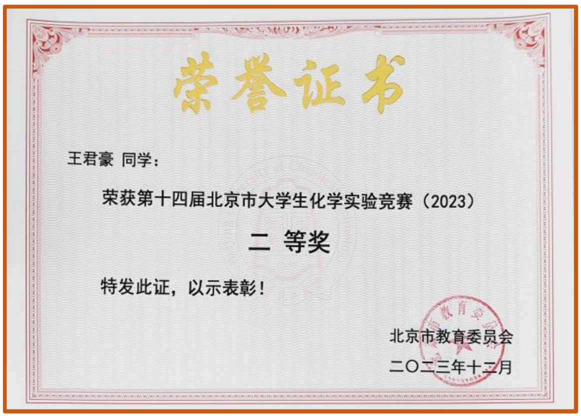

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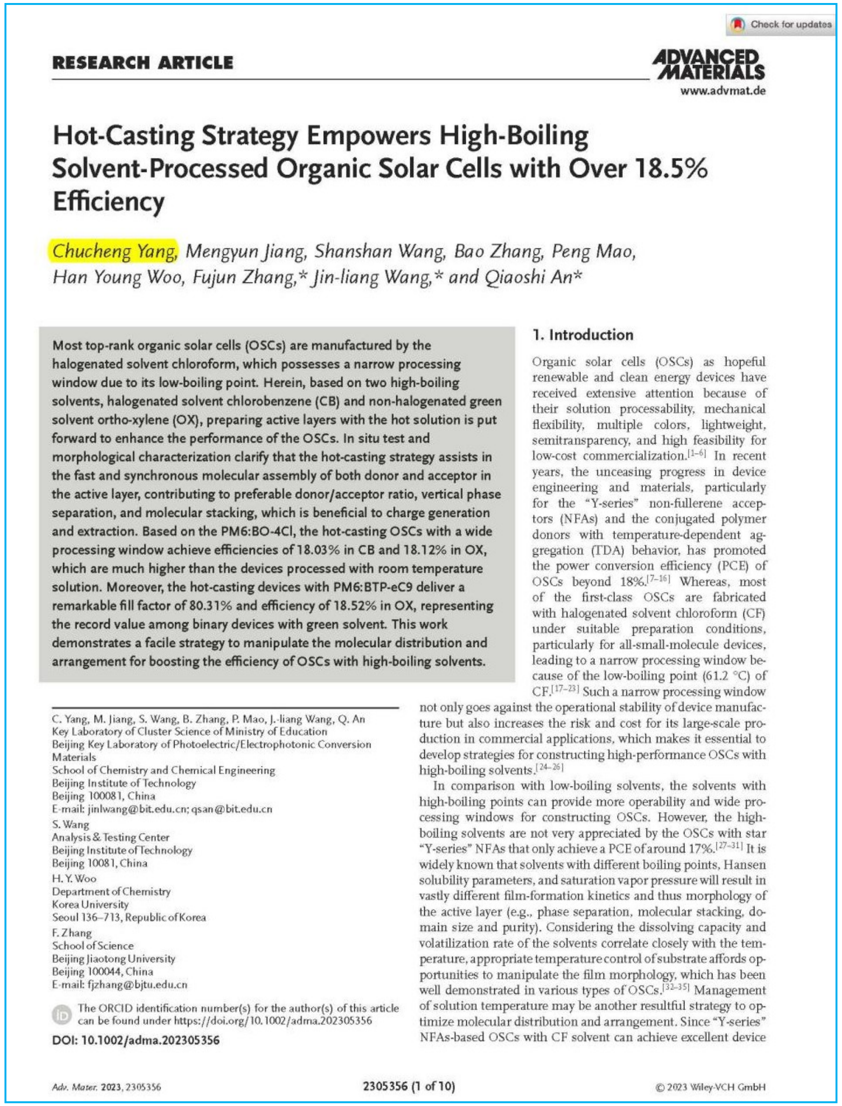
（2）代表性学术论文



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RESEARCH ARTICLE

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Hot-Casting Strategy Empowers High-Boiling Solvent-Processed Organic Solar Cells with Over 18.5% Efficiency

Chucheng Yang, Mengyun Jiang, Shanshan Wang, Bao Zhang, Peng Mao, Han Young Woo, Fujun Zhang, Jin-liang Wang,* and Qiaoshi An**

Most top-rank organic solar cells (OSCs) are manufactured by the halogenated solvent chloroform, which possesses a narrow processing window due to its low-boiling point. Herein, based on two high-boiling solvents, halogenated solvent chlorobenzene (CB) and non-halogenated green solvent ortho-xylene (OX), preparing active layers with the hot solution is put forward to enhance the performance of the OSCs. In situ test and morphological characterization clarify that the hot-casting strategy assists in the fast and synchronous molecular assembly of both donor and acceptor in the active layer, contributing to preferable donor/acceptor ratio, vertical phase separation, and molecular stacking, which is beneficial to charge generation and extraction. Based on the PM6:BO-4Cl, the hot-casting OSCs with a wide processing window achieve efficiencies of 18.03% in CB and 18.12% in OX, which are much higher than the devices processed with room temperature solution. Moreover, the hot-casting devices with PM6:BTP-eC9 deliver a remarkable fill factor of 80.31% and efficiency of 18.52% in OX, representing the record value among binary devices with green solvent. This work demonstrates a facile strategy to manipulate the molecular distribution and arrangement for boosting the efficiency of OSCs with high-boiling solvents.

1. Introduction

Organic solar cells (OSCs) as hopeful renewable and clean energy devices have received extensive attention because of their solution processability, mechanical flexibility, multiple colors, lightweight, semitransparency, and high feasibility for low-cost commercialization.[1–6] In recent years, the unceasing progress in device engineering and materials, particularly for the "Y-series" non-fullerene acceptors (NFAs) and the conjugated polymer donors with temperature-dependent aggregation (TDA) behavior, has promoted the power conversion efficiency (PCE) of OSCs beyond 18%.[7–16] Whereas, most of the first-class OSCs are fabricated with halogenated solvent chloroform (CF) under suitable preparation conditions, particularly for all-small-molecule devices, leading to a narrow processing window because of the low-boiling point (61.2 °C) of CF.[17–23] Such a narrow processing window not only goes against the operational stability of device manufacture but also increases the risk and cost for its large-scale production in commercial applications, which makes it essential to develop strategies for constructing high-performance OSCs with high-boiling solvents.[24–26]

In comparison with low-boiling solvents, the solvents with high-boiling points can provide more operability and wide processing windows for constructing OSCs. However, the high-boiling solvents are not very appreciated by the OSCs with star "Y-series" NFAs that only achieve a PCE of around 17%.[27–31] It is widely known that solvents with different boiling points, Hansen solubility parameters, and saturation vapor pressure will result in vastly different film-formation kinetics and thus morphology of the active layer (e.g., phase separation, molecular stacking, domain size and purity). Considering the dissolving capacity and volatilization rate of the solvents correlate closely with the temperature, appropriate temperature control of substrate affords opportunities to manipulate the film morphology, which has been well demonstrated in various types of OSCs.[32–35] Management of solution temperature may be another resultful strategy to optimize molecular distribution and arrangement. Since "Y-series" NFAs-based OSCs with CF solvent can achieve excellent device

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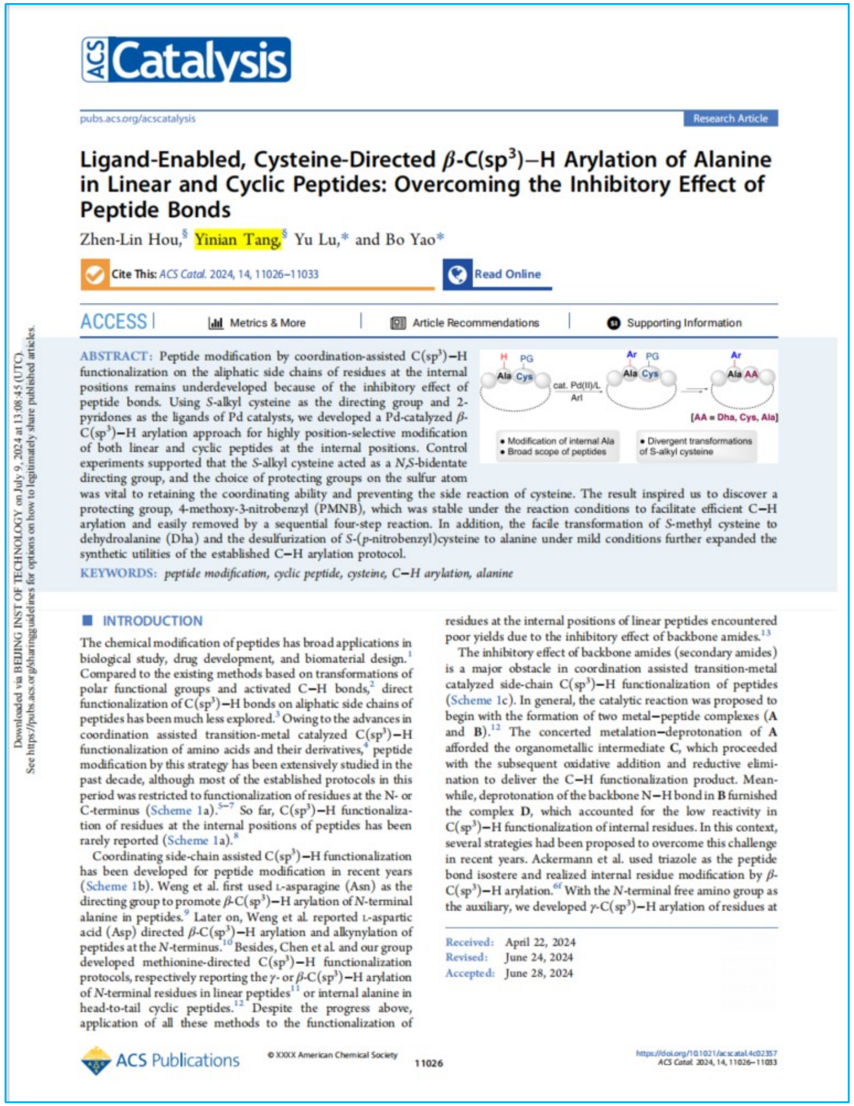

Catalysis

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Ligand-Enabled, Cysteine-Directed β-C(sp^3)−H Arylation of Alanine in Linear and Cyclic Peptides: Overcoming the Inhibitory Effect of Peptide Bonds

Zhen-Lin Hou,[§] Yinian Tang,[§] Yu Lu,* and Bo Yao*

Cite This: *ACS Catal.* 2024, 14, 11026−11033　　Read Online

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ABSTRACT: Peptide modification by coordination-assisted C(sp^3)−H functionalization on the aliphatic side chains of residues at the internal positions remains underdeveloped because of the inhibitory effect of peptide bonds. Using *S*-alkyl cysteine as the directing group and 2-pyridones as the ligands of Pd catalysts, we developed a Pd-catalyzed β-C(sp^3)−H arylation approach for highly position-selective modification of both linear and cyclic peptides at the internal positions. Control experiments supported that the *S*-alkyl cysteine acted as a *N,S*-bidentate directing group, and the choice of protecting groups on the sulfur atom was vital to retaining the coordinating ability and preventing the side reaction of cysteine. The result inspired us to discover a protecting group, 4-methoxy-3-nitrobenzyl (PMNB), which was stable under the reaction conditions to facilitate efficient C−H arylation and easily removed by a sequential four-step reaction. In addition, the facile transformation of *S*-methyl cysteine to dehydroalanine (Dha) and the desulfurization of *S*-(*p*-nitrobenzyl)cysteine to alanine under mild conditions further expanded the synthetic utilities of the established C−H arylation protocol.

KEYWORDS: *peptide modification, cyclic peptide, cysteine, C−H arylation, alanine*

INTRODUCTION

The chemical modification of peptides has broad applications in biological study, drug development, and biomaterial design.[1] Compared to the existing methods based on transformations of polar functional groups and activated C−H bonds,[2] direct functionalization of C(sp^3)−H bonds on aliphatic side chains of peptides has been much less explored.[3] Owing to the advances in coordination assisted transition-metal catalyzed C(sp^3)−H functionalization of amino acids and their derivatives,[4] peptide modification by this strategy has been extensively studied in the past decade, although most of the established protocols in this period was restricted to functionalization of residues at the N- or C-terminus (Scheme 1a).[5−7] So far, C(sp^3)−H functionalization of residues at the internal positions of peptides has been rarely reported (Scheme 1a).[8]

Coordinating side-chain assisted C(sp^3)−H functionalization has been developed for peptide modification in recent years (Scheme 1b). Weng et al. first used L-asparagine (Asn) as the directing group to promote β-C(sp^3)−H arylation of *N*-terminal alanine in peptides.[9] Later on, Weng et al. reported L-aspartic acid (Asp) directed β-C(sp^3)−H arylation and alkynylation of peptides at the *N*-terminus.[10] Besides, Chen et al. and our group developed methionine-directed C(sp^3)−H functionalization protocols, respectively reporting the γ- or β-C(sp^3)−H arylation of *N*-terminal residues in linear peptides[11] or internal alanine in head-to-tail cyclic peptides.[12] Despite the progress above, application of all these methods to the functionalization of residues at the internal positions of linear peptides encountered poor yields due to the inhibitory effect of backbone amides.[13]

The inhibitory effect of backbone amides (secondary amides) is a major obstacle in coordination assisted transition-metal catalyzed side-chain C(sp^3)−H functionalization of peptides (Scheme 1c). In general, the catalytic reaction was proposed to begin with the formation of two metal−peptide complexes (**A** and **B**).[12] The concerted metalation−deprotonation of **A** afforded the organometallic intermediate **C**, which proceeded with the subsequent oxidative addition and reductive elimination to deliver the C−H functionalization product. Meanwhile, deprotonation of the backbone N−H bond in **B** furnished the complex **D**, which accounted for the low reactivity in C(sp^3)−H functionalization of internal residues. In this context, several strategies had been proposed to overcome this challenge in recent years. Ackermann et al. used triazole as the peptide bond isostere and realized internal residue modification by β-C(sp^3)−H arylation.[6f] With the *N*-terminal free amino group as the auxiliary, we developed γ-C(sp^3)−H arylation of residues at

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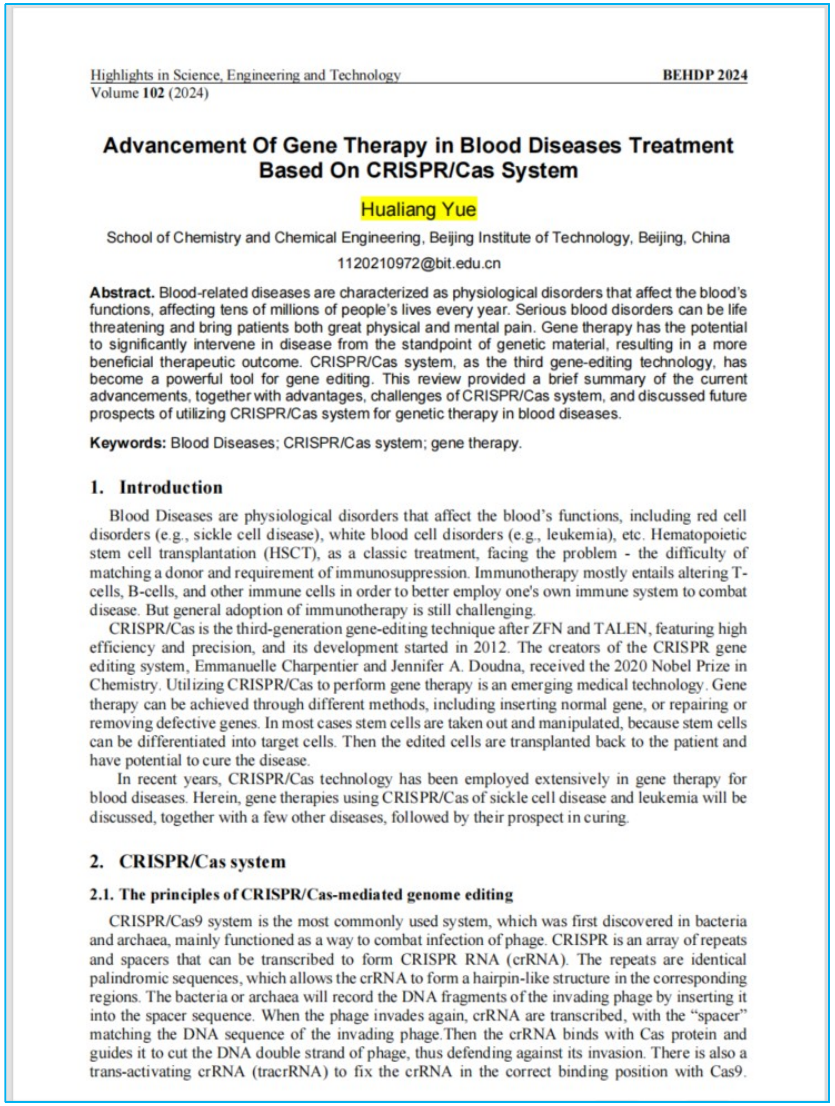

Highlights in Science, Engineering and Technology BEHDP 2024
Volume **102** (2024)

Advancement Of Gene Therapy in Blood Diseases Treatment Based On CRISPR/Cas System

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Abstract. Blood-related diseases are characterized as physiological disorders that affect the blood's functions, affecting tens of millions of people's lives every year. Serious blood disorders can be life threatening and bring patients both great physical and mental pain. Gene therapy has the potential to significantly intervene in disease from the standpoint of genetic material, resulting in a more beneficial therapeutic outcome. CRISPR/Cas system, as the third gene-editing technology, has become a powerful tool for gene editing. This review provided a brief summary of the current advancements, together with advantages, challenges of CRISPR/Cas system, and discussed future prospects of utilizing CRISPR/Cas system for genetic therapy in blood diseases.

Keywords: Blood Diseases; CRISPR/Cas system; gene therapy.

1. Introduction

Blood Diseases are physiological disorders that affect the blood's functions, including red cell disorders (e.g., sickle cell disease), white blood cell disorders (e.g., leukemia), etc. Hematopoietic stem cell transplantation (HSCT), as a classic treatment, facing the problem - the difficulty of matching a donor and requirement of immunosuppression. Immunotherapy mostly entails altering T-cells, B-cells, and other immune cells in order to better employ one's own immune system to combat disease. But general adoption of immunotherapy is still challenging.

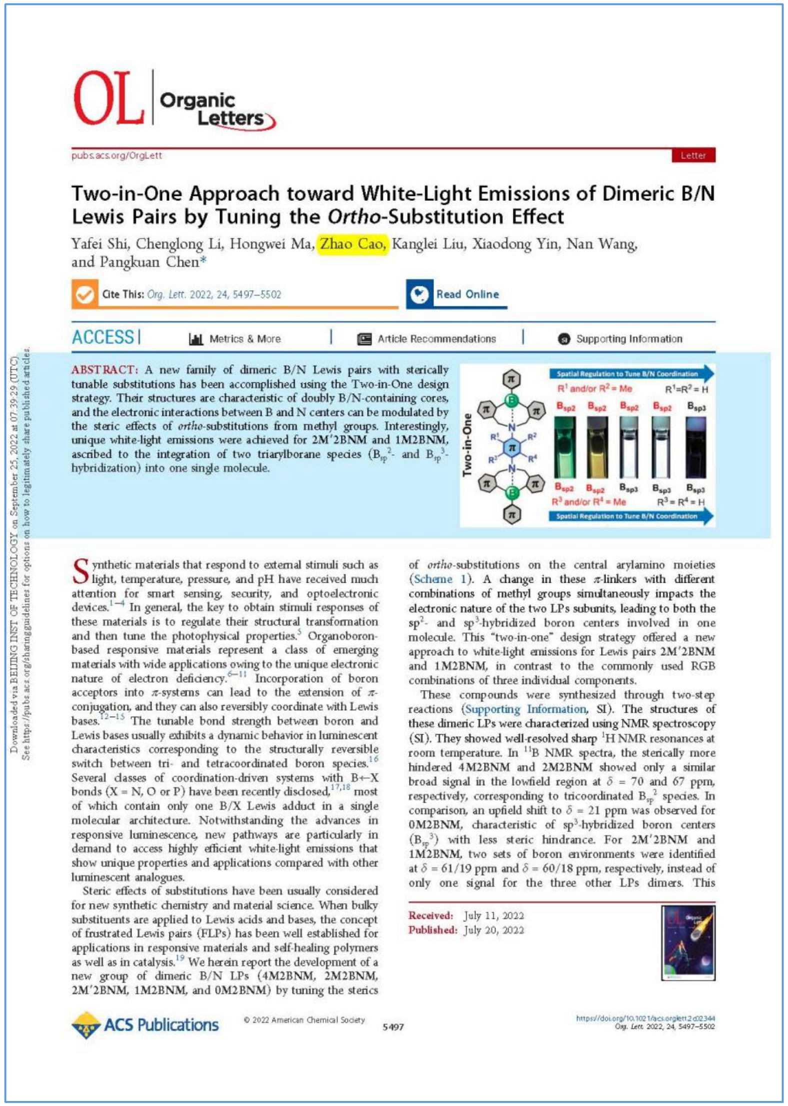
CRISPR/Cas is the third-generation gene-editing technique after ZFN and TALEN, featuring high efficiency and precision, and its development started in 2012. The creators of the CRISPR gene editing system, Emmanuelle Charpentier and Jennifer A. Doudna, received the 2020 Nobel Prize in Chemistry. Utilizing CRISPR/Cas to perform gene therapy is an emerging medical technology. Gene therapy can be achieved through different methods, including inserting normal gene, or repairing or removing defective genes. In most cases stem cells are taken out and manipulated, because stem cells can be differentiated into target cells. Then the edited cells are transplanted back to the patient and have potential to cure the disease.

In recent years, CRISPR/Cas technology has been employed extensively in gene therapy for blood diseases. Herein, gene therapies using CRISPR/Cas of sickle cell disease and leukemia will be discussed, together with a few other diseases, followed by their prospect in curing.

2. CRISPR/Cas system

2.1. The principles of CRISPR/Cas-mediated genome editing

CRISPR/Cas9 system is the most commonly used system, which was first discovered in bacteria and archaea, mainly functioned as a way to combat infection of phage. CRISPR is an array of repeats and spacers that can be transcribed to form CRISPR RNA (crRNA). The repeats are identical palindromic sequences, which allows the crRNA to form a hairpin-like structure in the corresponding regions. The bacteria or archaea will record the DNA fragments of the invading phage by inserting it into the spacer sequence. When the phage invades again, crRNA are transcribed, with the "spacer" matching the DNA sequence of the invading phage.Then the crRNA binds with Cas protein and guides it to cut the DNA double strand of phage, thus defending against its invasion. There is also a trans-activating crRNA (tracrRNA) to fix the crRNA in the correct binding position with Cas9.



OL | Organic Letters
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Two-in-One Approach toward White-Light Emissions of Dimeric B/N Lewis Pairs by Tuning the *Ortho*-Substitution Effect

Yafei Shi, Chenglong Li, Hongwei Ma, Zhao Cao, Kanglei Liu, Xiaodong Yin, Nan Wang, and Pangkuan Chen*

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ABSTRACT: A new family of dimeric B/N Lewis pairs with sterically tunable substitutions has been accomplished using the Two-in-One design strategy. Their structures are characteristic of doubly B/N-containing cores, and the electronic interactions between B and N centers can be modulated by the steric effects of *ortho*-substitutions from methyl groups. Interestingly, unique white-light emissions were achieved for 2M′2BNM and 1M2BNM, ascribed to the integration of two triarylborane species ($B_{sp}{}^{2}$- and $B_{sp}{}^{3}$-hybridization) into one single molecule.

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Synthetic materials that respond to external stimuli such as light, temperature, pressure, and pH have received much attention for smart sensing, security, and optoelectronic devices.[1−4] In general, the key to obtain stimuli responses of these materials is to regulate their structural transformation and then tune the photophysical properties.[5] Organoboron-based responsive materials represent a class of emerging materials with wide applications owing to the unique electronic nature of electron deficiency.[6−11] Incorporation of boron acceptors into π-systems can lead to the extension of π-conjugation, and they can also reversibly coordinate with Lewis bases.[12−15] The tunable bond strength between boron and Lewis bases usually exhibits a dynamic behavior in luminescent characteristics corresponding to the structurally reversible switch between tri- and tetracoordinated boron species.[16] Several classes of coordination-driven systems with B←X bonds (X = N, O or P) have been recently disclosed,[17,18] most of which contain only one B/X Lewis adduct in a single molecular architecture. Notwithstanding the advances in responsive luminescence, new pathways are particularly in demand to access highly efficient white-light emissions that show unique properties and applications compared with other luminescent analogues.

Steric effects of substitutions have been usually considered for new synthetic chemistry and material science. When bulky substituents are applied to Lewis acids and bases, the concept of frustrated Lewis pairs (FLPs) has been well established for applications in responsive materials and self-healing polymers as well as in catalysis.[19] We herein report the development of a new group of dimeric B/N LPs (4M2BNM, 2M2BNM, 2M′2BNM, 1M2BNM, and 0M2BNM) by tuning the sterics of *ortho*-substitutions on the central arylamino moieties (Scheme 1). A change in these π-linkers with different combinations of methyl groups simultaneously impacts the electronic nature of the two LPs subunits, leading to both the sp^2- and sp^3-hybridized boron centers involved in one molecule. This "two-in-one" design strategy offered a new approach to white-light emissions for Lewis pairs 2M′2BNM and 1M2BNM, in contrast to the commonly used RGB combinations of three individual components.

These compounds were synthesized through two-step reactions (Supporting Information, SI). The structures of these dimeric LPs were characterized using NMR spectroscopy (SI). They showed well-resolved sharp ^{1}H NMR resonances at room temperature. In ^{11}B NMR spectra, the sterically more hindered 4M2BNM and 2M2BNM showed only a similar broad signal in the lowfield region at $\delta = 70$ and 67 ppm, respectively, corresponding to tricoordinated $B_{sp}{}^{2}$ species. In comparison, an upfield shift to $\delta = 21$ ppm was observed for 0M2BNM, characteristic of sp^3-hybridized boron centers ($B_{sp}{}^{3}$) with less steric hindrance. For 2M′2BNM and 1M2BNM, two sets of boron environments were identified at $\delta = 61/19$ ppm and $\delta = 60/18$ ppm, respectively, instead of only one signal for the three other LPs dimers. This

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