

## Synthetic Methods

Heteroatom Cycloaddition at the (BN)<sub>2</sub> Bay Region of Dibenzoperylene

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**Abstract:** Cycloaddition-dehydration involving a BNBN-butadiene analogue at the bay region of a dibenzoperylene and a non-enolizable aldehyde provides a novel strategy for incorporation of the oxadiazaborinane (B<sub>2</sub>N<sub>2</sub>CO) ring into the scaffold of a polycyclic aromatic hydrocarbon resulting in highly emissive compounds.

The introduction of heteroatoms is a very popular strategy to modify the electronic properties of polycyclic aromatic hydrocarbons (PAHs) for application as organic electronic materials.<sup>[1]</sup> In particular, the isoelectronic and isosteric exchange of carbon-carbon (CC) by boron and nitrogen pairs (BN) opened a huge field of novel organic materials since the report of the first BN doped PAH in 1958.<sup>[2]</sup> In addition to BN substitution, the incorporation of oxygen atoms is a useful approach to develop new compounds for material applications and catalysis.<sup>[1,3]</sup>

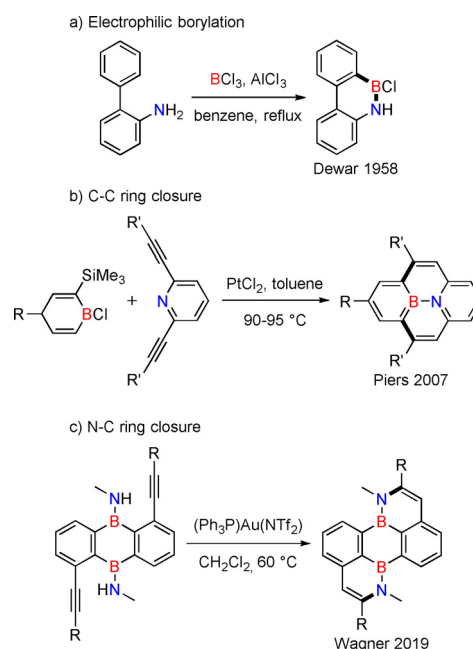
Among the available methods for synthesizing 1,2-azaborine containing PAHs, the electrophilic borylation, introduced by Dewar<sup>[2a]</sup> and optimized further, is attractive as both B–N and B–C bonds are constructed in one synthetic step (Scheme 1 a).<sup>[2h,3a,4]</sup> Alternatively, the coupling of carbon-carbon (CC) bonds around existing BN units has occasionally been employed, e.g., by Piers,<sup>[5]</sup> Bonifazi,<sup>[3b,6]</sup> and our group (Scheme 1 b).<sup>[7]</sup> The Wagner group<sup>[8]</sup> described the Au catalyzed NC bond formation as the final step of the synthesis of a doubly BN substituted perylene (Scheme 1 c).<sup>[8,9]</sup>

Perylenes and dibenzoperylenes with BN doping have been synthesized by Wagner, Bonifazi and our group.<sup>[3b,4j,8,10]</sup> We have developed a multistep synthesis of a dibenzoperylene

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**Scheme 1.** Selected representative examples of available methods for synthesis of 1,2-azaborine containing PAHs.

**1** that features a BNBN unit at its bay region.<sup>[4j]</sup> It would be interesting to use this heterodiene as functional group for widening the chemical space in heteroatom doped PAH synthesis by cycloaddition reactions. Cycloaddition reactions at the bay region of perylene are well known (Scheme 2),<sup>[11]</sup> and they were even suggested for growth of carbon nanotubes by reaction of armchair rims with acetylene.<sup>[12]</sup> However, the analogous cycloaddition reaction of heteroatom bay region substituted PAHs are unknown. The closest to this are cycloaddition reactions of diazadiboretidines, which are the quite reactive BN analogs of antiaromatic cyclobutadiene. Paetzold postulated hetero-Diels–Alder reactions of diazadiboretidines with ketones and aldehydes forming 1,3,5,2,4-oxadiazaborinane (B<sub>2</sub>N<sub>2</sub>CO) rings under ring extension (Scheme 2).<sup>[13]</sup>

The cycloaddition of **A** (a model for **1** without the n-Bu groups) with typical electron rich (TCNE, TME) or electron poor dienophiles (methyl vinyl ether, see Figure 1) is unfavorable as revealed by quantum chemical calculations (M062X/6–311 + G\*\*), even after dehydration. However, the reaction **A** → **C** + H<sub>2</sub>O becomes mildly exergonic if the polar carbonyl group of benzaldehyde is employed as

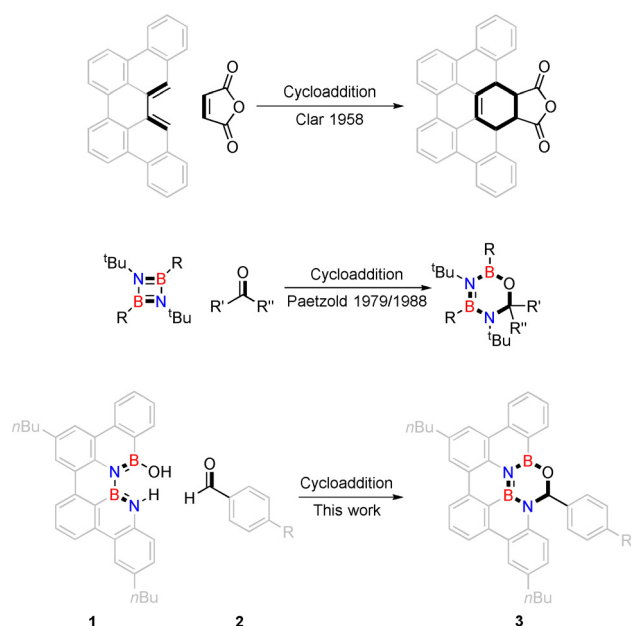
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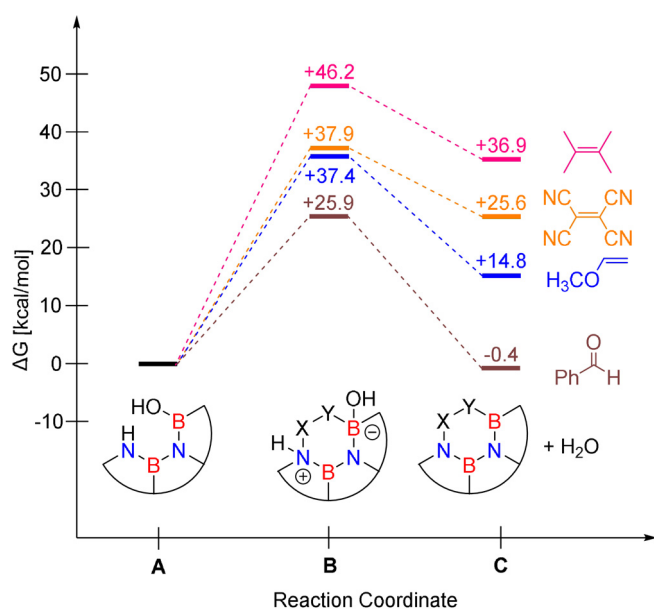
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**Scheme 2.** Cycloaddition reactions of dibenzoperylene (top), diazadiboretidines (center) and BN bay region doped dibenzoperylene (bottom).



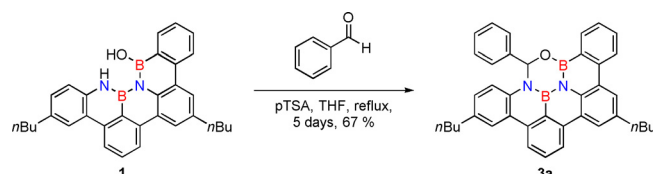
**Figure 1.** Free energy differences for different dienophiles calculated at the M062X/6-311+G\*\* level of theory ( $T=298.15$  K); (BN)<sub>2</sub> dibenzoperylene scaffolds **A**, **B** and **C** are abbreviated for clarity.

dienophile. For reactions with other hetero dienophiles, see Supporting Information (SI).

Heating **1** and benzaldehyde in THF even for a long period of time does not result in any reaction. To facilitate the dehydration that is necessary to have an exergonic reaction, we added an excess of para-toluenesulfonic acid (pTSA) in order to induce an acid catalyzed dehydration. To remove emerging water from equilibrium, we heated the mixture in the presence of molecular sieve in a Soxhlet extractor. Then, a slow reaction proceeds and after 5 days of reaction time, the

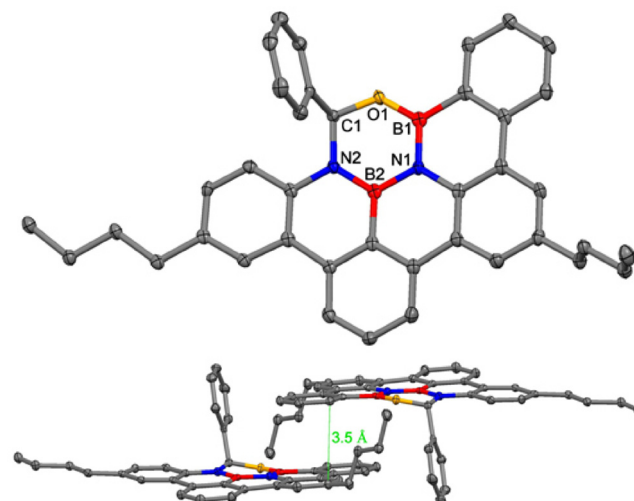
product **3a**, which is stable against oxygen and moisture, can be isolated in a good yield of 67% by column chromatography.

Compound **3a** (Scheme 3) can be characterized by multinuclear (<sup>1</sup>H, <sup>13</sup>C, <sup>11</sup>B) and correlated NMR spectroscopy (2D spectra) as well as by high resolution mass spectrometry. Notably, the <sup>1</sup>H NMR shows one singlet for the benzylic proton in the ON<sub>2</sub>B<sub>2</sub>C-ring that experiences significant deshielding and resonates at 7.02 ppm (see SI, Figure S1). The <sup>11</sup>B spectrum of **3a** shows a broad signal at 28.3 ppm for both boron atoms (see SI, Figure S3).



**Scheme 3.** Heteroatom cycloaddition reaction with benzaldehydes; 1 equiv **1**, 10 equiv benzaldehyde, 10 equiv pTSA, THF, Soxhlet extractor with 3 Å molecular sieve, reflux, 5 days.

Single crystals for X-ray crystallography were obtained by recrystallization from THF (for crystallographic data see SI). Compound **3a** crystallizes in the triclinic space group  $P\bar{1}$  with two enantiomeric molecules due to the asymmetric sp<sup>3</sup> carbon in the B<sub>2</sub>N<sub>2</sub>CO ring (see Figure 2). The dibenzoperylene motif is responsible for intermolecular  $\pi$ - $\pi$  stacking with a distance of 3.5 Å between molecular planes, while the single phenyl groups as well as one butyl group feature towards the next molecular layer. The BN bond lengths average 1.43 Å, slightly shorter than analogous bond lengths in typical BN doped PAHs (1.45–1.47 Å).<sup>[5a,14]</sup> The CO, BO and NC bond length are in good agreement with the literature.<sup>[15]</sup> The newly formed heterocycle adopts a puckered geometry with a tilt angle of 18.15° involving the BNB and NCO planes. This ring



**Figure 2.** Molecular structure of **3a** (hydrogen atoms omitted for clarity (top), ellipsoids set at 50% probability; selected bond length of **3a** in Å: O1-B1 1.376, B1-N1 1.420, N1-B2 1.444, B2-N2 1.422, N2-C1 1.468, C1-O1 1.430; packing of **3a** (bottom).<sup>[20]</sup>

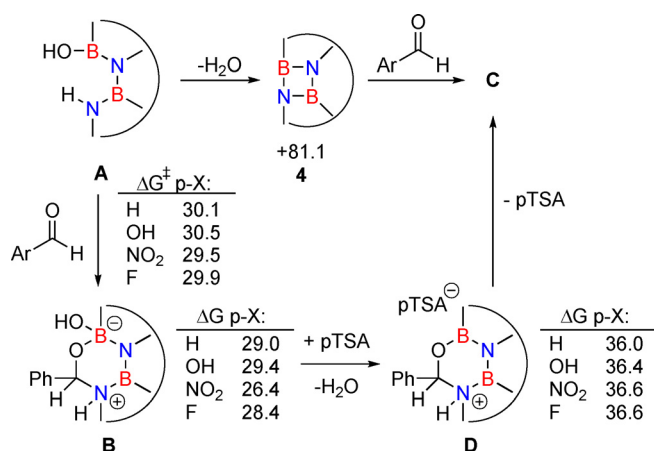
has a  $sp^3$  hybridized carbon center and is expected to be non-aromatic as confirmed by NICS(1) computations (see SI, Figure S40).<sup>[16]</sup> While the four all-carbon rings of the PAH backbone have large negative NICS(1), the heteroatom containing rings have small NICS(1) values, reminiscent of the related boroxazine derivative.<sup>[3c]</sup>

In order to investigate substituent effects, a number of aryl aldehydes were employed in the reaction (Table 1). The reaction time and yield are not affected significantly by the electronic nature of the substituent (Table 1, entries 1–4) as revealed by TLC monitoring of the disappearance of **1**. Mesitaldehyde can also be used as a dienophile, but provides lower yields due to steric hindrance (Table 1, entry 5). The product with the sterically more encumbered pivalaldehyde can only be detected in low yields (Table 1, entry 6) and with some impurities (see SI Figure S32). The  $\alpha,\beta$ -unsaturated cinnamaldehyde undergoes this cycloaddition reaction in good yields (Table 1, entry 7) and selectively at the aldehyde function as no product resulting from cycloaddition to the  $C=C$  double bond is observed. All dehydrated cycloaddition products are stable against oxygen and moisture and can be purified by column chromatography.

**Table 1:** Screening of different dienophiles in cycloaddition reaction with  $(BN)_2$ -dibenzoperylene and yield values. Conditions as in Scheme 3.

Entry	Dienophile	Product	Yield [%]
1			67
2			74
3			73
4			80
5			40
6			15
7			77

The influence of pTSA on the reagents was investigated by NMR spectroscopy in  $[D_8]THF$ . A mixture of pTSA and benzaldehyde gives unchanged  $^1H$  NMR spectra compared to individual compounds. This observation is in line with the  $pK_a$  values of pTSA ( $-2.8$ )<sup>[17]</sup> and of protonated benzaldehyde ( $-7.1$ ).<sup>[18]</sup> On the other hand, the signal of the OH group of **1** at 9.06 ppm (assignment based on 2D NMR) turns broad at low concentrations of pTSA and becomes weaker and broader with increasing the pTSA concentration, indicating rapid exchange between the OH proton and the acidic proton of pTSA. The NH proton is not affected by the presence of pTSA and its signal remains sharp at 9.78 ppm. These results indicate that the OH group is involved in dynamic proton exchange with pTSA in THF solution. The action of pTSA could result in acid catalyzed dehydration of **1** to give the diazadiboretidine intermediate **4** (Scheme 4). Recall that



**Scheme 4.** Possible mechanisms as computed at the M062X/6-311+G\*\* level of theory in THF solution ( $T=339$  K). Energy data is given in  $kcal\ mol^{-1}$ .

these species were previously reported to undergo cycloaddition reactions with aldehydes (see Scheme 2).<sup>[13a,b]</sup> Computations on the model system **A** without the n-Bu groups at the M062X/6-311+G\*\* level of theory with a continuum model to mimic the THF solvent effect show that formation of **4** is a very high energy process,  $\Delta G(THF) = +81.1\ kcal\ mol^{-1}$  (Scheme 4). Therefore, **4** cannot be relevant for the mechanism of the dehydrative cycloaddition reaction. However, neutral **A** can undergo the concerted cycloaddition with benzaldehyde with a barrier of  $30.1\ kcal\ mol^{-1}$ , and this barrier is changed by at most  $0.4\ kcal\ mol^{-1}$  in the presence of para substituents (p-OH, p- $NO_2$ , p-F) on the aryl group of the aldehyde (Scheme 4). The cycloaddition product **B** is a very shallow minimum on the potential energy surface as the barrier for the back reaction to the reagents is merely 1–3  $kcal\ mol^{-1}$  depending on p-X (Scheme 4). The pTSA could then cause acid catalyzed dehydration of **B** to give intermediate **D** (Scheme 4) that is higher in energy than **B** by roughly 8  $kcal\ mol^{-1}$ . The energy barriers of the cycloaddition step and its insensitivity to electronic substituent effects of the aryl aldehydes are in qualitative agreement with the experimental observations that the reaction rate is independent of

the benzaldehyde concentration but that addition of acid is essential. Alternatively, the OH protonated form of **1**, a borenium ion, could undergo the cycloaddition presumably with a decreased activation barrier, but the Gibbs free energy ( $T = 339\text{ K}$ ) of this borenium ion is higher than that of **D** by roughly  $12\text{ kcal mol}^{-1}$ .

The absorption and fluorescence spectra of compounds **3a–e** and **3g** were recorded in dichloromethane solutions (see Figure 3). The spectra resemble each other and are quite similar to those of **1**. The substituents in **3b–e** or the presence of the C=C bridge in **3g** turned out to result in shifts of peak maxima of 1–2 nm. The Stokes shifts are small throughout ( $600\text{ cm}^{-1}$ – $755\text{ cm}^{-1}$ , see SI for details). The fluorescence quantum yields of all products except for **3c** is 78% and higher using 9,10-diphenylanthracene in ethanol as reference (see SI). The cycloaddition product with p-nitrobenzaldehyde **3c** only shows a low fluorescence quantum yield of 7%. The decrease of the fluorescence quantum yield of aromatic compounds upon introduction of a nitro groups is well known.<sup>[19]</sup>

The electrochemical properties of **3a** were investigated by cyclic voltammetry. At scan rates greater than  $2\text{ V s}^{-1}$ , we find a quasi-reversible reduction signal at  $E^0 = -2.724\text{ V vs. Fc/}$

$\text{Fc}^+$ . At slow scan rates, the associated oxidation peak of this reduction signal is less pronounced, suggesting a slow chemical follow-up reaction of the reduced species of **3a** (EC mechanism). Two further signals with peak potentials at approx.  $-3.0\text{ V}$  and  $-3.1\text{ V vs. Fc/}$  were found in the forward scan at  $50\text{ mV s}^{-1}$  within our electrochemical window (see Supporting Information for details).

In summary, we described the first cycloaddition-dehydration reaction of a BNB bay region doped dibenzoperylene with different non-enolizable aldehydes forming new BN doped PAHs with an 1,3,5,2,4-oxadiazadiborinan ( $\text{B}_2\text{N}_2\text{CO}$ ) ring. The reaction is robust towards the electronic structure of the aldehydes, but is affected by steric effects. The novel reaction products, PAH with an embedded oxadiazadiborinane ring, show very high fluorescence quantum yields ( $>78\%$ ), which make this conjugation of BN-doped PAH with aldehydes a promising strategy towards enlarging the chemical scope in heteroatom doped PAH chemistry.

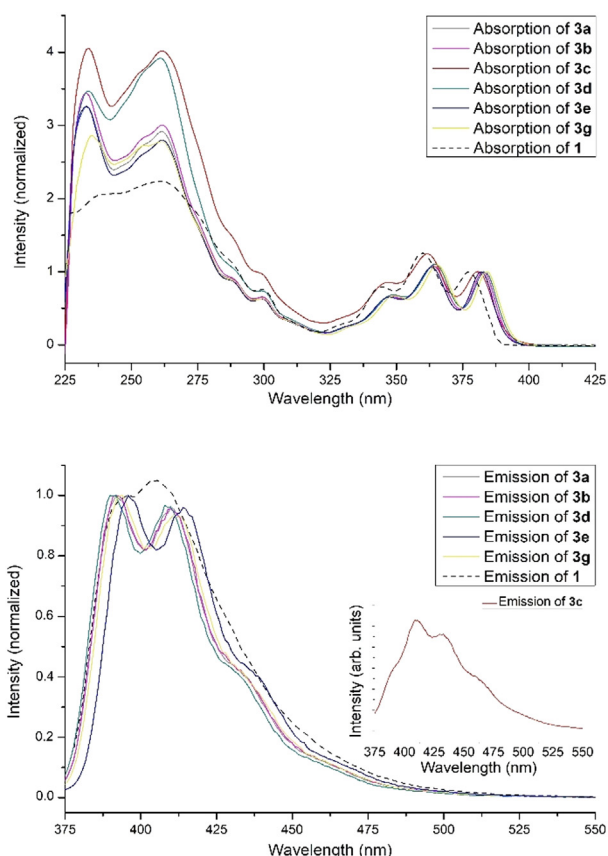
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### Conflict of interest

The authors declare no conflict of interest.

**Keywords:** boron · cycloaddition · dibenzoperylene · nitrogen · polycycles



**Figure 3.** Absorption (normalized on maxima with longest wavelength, top) and fluorescence spectra ( $\lambda_{\text{ex}} = 347\text{ nm}$ , normalized on maxima with shortest wavelength, bottom) of the cycloaddition products in dichloromethane including absorption and fluorescence spectra of **1** (dashed line) for comparison. Inset: fluorescence spectrum of **3c** ( $\Phi_{\text{fl}} = 0.07$ ).

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