








# A simplified approach to thermally activated delayed fluorescence (TADF) bipolar host polymers†

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Organic Light Emitting Diodes (OLEDs) are a critical part of current consumer electronics, from mobile device displays to solid-state lighting. Thus demand for the development of industrially compatible OLED materials continues to increase. Herein, we introduce a series of solution-processible polymers incorporating a Thermally Activated Delayed Fluorescence (TADF) emitter and a host species in the side chain to finely adjust the charge transport properties. For balanced charge transport, a bipolar host polymer based on carbazole and  $\alpha$ -carboline was investigated in addition to the commonly employed unipolar host mCP and an electron transporting version thereof. We demonstrate that the combination of unipolar co-hosts on one polymer chain can generate, with less synthetic effort, the same optoelectronic properties as a polymer carrying the corresponding bipolar host molecule as a pendant.

## Introduction

Organic Light Emitting Diodes (OLEDs) are currently surpassing other technologies in mobile device displays, large area TVs and solid-state lighting for reasons including lower energy consumption and the possibility of lighter-weight, flexible devices.<sup>1</sup> Additionally, fabrication methods for OLEDs are more scalable and cost-efficient due to their solution-based nature. After fluorescent and phosphorescent emitter materials,<sup>2</sup> a new class of emitters, based on Thermally Activated Delayed Fluorescence (TADF)<sup>3</sup> has emerged over the last decade using heavy metal-

free organic molecules that harvest singlet as well as triplet excitons and thus can theoretically reach 100% internal quantum efficiencies through delayed fluorescence.<sup>4</sup>

However, to date most of the OLED emissive layers (EML) consist of small molecule emitters doped into a host matrix which is only processable by evaporation techniques.<sup>5</sup> Since the pioneering work of Burroughes *et al.* in 1990<sup>6</sup> when reporting the first polymer light emitting diode, a number of dendrimers and polymers have been investigated in place of small molecules in the quest to develop more efficient emitter materials. Devices based on these high molecular weight materials can be fabricated by precise deposition methods such as the industrially favoured ink-jet printing.

Nevertheless, most research efforts to date focused on conjugated polymers as the active emitter,<sup>7</sup> which can come with tedious monomer synthesis and complicated polymerization procedures that cannot be easily achieved with higher polymerization degrees. Moreover, the electronic properties of conjugated polymers strongly depend on the molecular weight and defects along the polymer chain and hence are difficult to reproduce.<sup>8</sup> A common design strategy for TADF polymers is the incorporation of the TADF unit along the conjugated<sup>9</sup> or non-conjugated<sup>10</sup> polymer backbone. In general, the TADF emitter is added as an intact unit in the side chain,<sup>11</sup> although other approaches<sup>12</sup> found that the TADF property can be generated through space charge transfer of vicinal donor and acceptor side chain units.<sup>13</sup>

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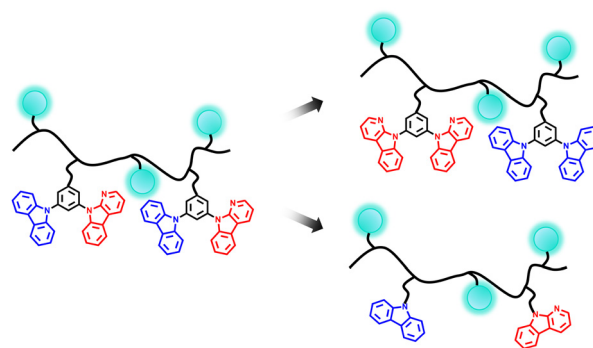
†Electronic supplementary information (ESI) available: Synthetic and experimental procedures, <sup>1</sup>H-NMR-spectra, SEC and CV data. See DOI: <https://doi.org/10.1039/d2py00511e>

Nonetheless, many systems do not comprise a host material and still require the addition of a small molecule host species in the OLED active emissive layer to achieve good results.<sup>14</sup> The addition of small molecules is not favourable for solution-processing as it changes the ink viscosity and influences the film formation. Host molecules, however, play a critical role in the EML to prevent concentration quenching by providing a matrix in which the emitter dopant is dispersed with aligned frontier orbital levels for good charge transport and high triplet energy to suppress exciton trapping. For example, a commercial host that is often used in blue and green phosphorescent and TADF OLEDs is 1,3-bis(*N*-carbazolyl)benzene (mCP) with good hole conductivity and a high triplet energy of 2.91 eV.<sup>15</sup>

Even though many host molecules are either p-type (hole-transporting) or n-type (electron-transporting) materials, a balanced transport of positive and negative charges in the EML is critical for a high recombination efficiency and external quantum efficiency (EQE). To broaden the charge recombination zone in the EML, bipolar self-hosted materials are designed to include electron- as well as hole-transporting moieties in their chemical structure.<sup>16</sup> Among the latter, carbazole is the most commonly incorporated conjugated moiety due to its high triplet energy, moderate hole transport properties, high chemical and thermal stability as well as simple synthetic modification.<sup>17</sup> The corresponding electron-accepting counterpart is pyridoindol, better known as carboline, with an additional nitrogen atom at the  $\alpha$ -,  $\beta$ - or  $\gamma$ -position.<sup>18</sup> Carbazole and carboline have been included in various combinations to form bipolar host materials that are suitable for (deep) blue and green OLED devices.<sup>19</sup> Among those, Lee and coworkers studied the bipolar version of mCP in which one carbazole is replaced by different carbolines and found that the  $\alpha$ -substitution in CzCb1 showed superior performance.<sup>20</sup> Further, these authors investigated the mCP derivative NmCP1 containing two carboline units instead of carbazole, where again the  $\alpha$ -substituted material delivered the best device results.<sup>21</sup>

In our previous work,<sup>22</sup> we designed a methacrylate based comonomer carrying the host material mCP as a pendant group, which enables the generation of an emissive copolymer with the TADF emitter 4-(9*H*-carbazol-9-yl)-2-(3'-hydroxy-[1,1'-biphenyl]-3-yl)-isoindoline-1,3-dione upon simple free radical polymerization.<sup>23</sup> In this approach, all the active components of the EML are connected by an inactive polymer backbone to form a one component system which can be easily solution-processed or even encapsulated in a semi-interpenetrating polymer network to enable multilayer solution-processing.<sup>22</sup>

Herein, we investigate the influence of the host species on the TADF emissive copolymer based on carbazole and  $\alpha$ -carboline as well as the hole and electron transporting derivatives of mCP. Furthermore, we contrast three approaches to incorporate the host material to obtain a balanced charge transport in the emissive copolymer (Scheme 1). Firstly, the asymmetric version of mCP with bipolar charge transporting properties, CzCb1, is functionalized as a methacrylate and copolymerized with the emitter. Secondly, in a novel approach to



**Scheme 1** Overview of the approach for simplified bipolar self-hosted TADF copolymers (the TADF emitter is depicted as light blue spheres, hole-transporting structural motives of the host species in blue, electron-transporting structural motives of the host species in red). A non-conjugated copolymer carrying a TADF emitter and a bipolar host CzCb1 (left) is contrasted with a TADF copolymer decorated with two unipolar co-hosts based on mCP (top right) and a further simplified version utilizing carbazole and  $\alpha$ -carboline (bottom right) as unipolar charge transporting species.

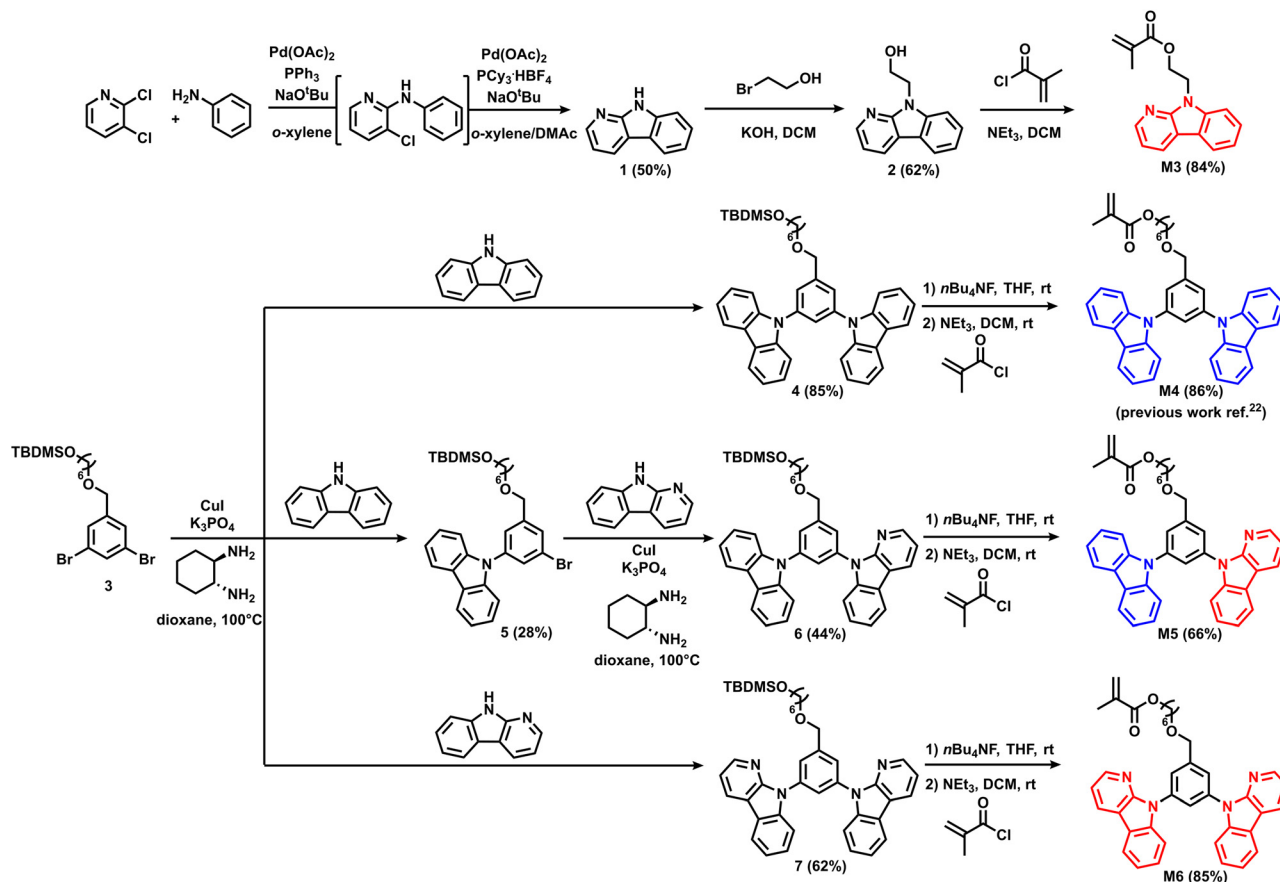
obtain bipolar charge transport, a combination of the unipolar hole and electron transporting co-hosts were copolymerized in an equal ratio. The advantage of this strategy is a simple adjustment and equilibration of hole and electron transporting species through the monomer ratio as well as the avoidance of the generally low yielding asymmetric synthesis which, additionally, constitutes more synthetic effort. Lastly, to further simplify the (monomer) synthesis, a copolymer using only carbazole and  $\alpha$ -carboline as pendant groups was investigated.

## Results and discussion

### Monomer synthesis

Whereas the synthesis of the  $\alpha$ -carboline methacrylate monomer **M3** (Scheme 2) has not previously been reported, the synthesis of the methacrylate monomer **M2** carrying carbazole as a side chain is well known in literature.<sup>24</sup> Based on this procedure,  $\alpha$ -carboline, generated in a palladium catalysed one-pot synthesis according to Cuny and coworkers,<sup>25</sup> was subsequently substituted at the N-H position with bromoethanol to yield 62% product. In a final esterification, the carboline methacrylate monomer **M3** was obtained in a high yield.

Previously, we reported a five-step synthesis route to obtain a methacrylate with mCP as a pendant group in good yields, in which first a linker benzene **3** is constructed, then the mCP unit **4** is formed and finally the methacrylate function introduced into the final molecule **M4** (Scheme 2).<sup>22</sup> Starting from the protected linker dibromobenzene **3**, the bipolar CzCb1 methacrylate can be obtained by a copper catalyzed Ullman coupling in an asymmetric substitution of the dibromobenzene **3** first with one carbazole group and subsequently with  $\alpha$ -carboline. However, in the first substitution with carbazole to obtain **5**, the generation of the twice substituted side product **4** can hardly be suppressed even if only small portions



**Scheme 2** Synthetic overview with experimental conditions for methacrylate monomers **M3**, **M4**, **M5** and **M6** carrying the respective host species as pendant. The hole-transporting species are marked in blue and the electron-transporting in red.

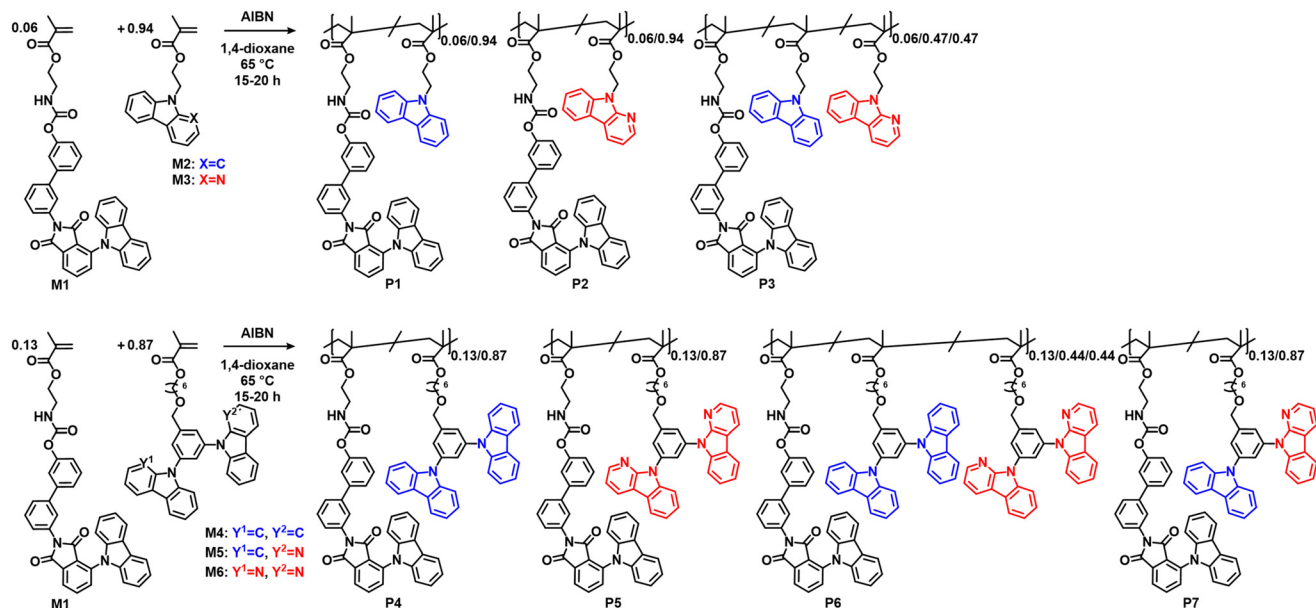
of carbazole are added at a time. To avoid the challenging separation of the side products *via* column chromatography, the reaction was stopped at lower conversions of *ca.* 40% prior to side product formation, yielding 28% of the pure monosubstituted product **5** while the starting material **3** could be recovered. The next step is the aromatic substitution with  $\alpha$ -carboline to build the bipolar aromatic system. Compared to carbazole,  $\alpha$ -carboline is a weaker nucleophile and even with an excess of reagent modest yields of 44% were achieved for the second substitution. For the unipolar NmCP1 unit **7**, the double substitution product was isolated in 62% yield. Overall, the asymmetric synthesis of the aromatic unit for the bipolar CzCb1 methacrylate yields only 12%, whereas the symmetric substitution to obtain the unipolar mCP achieves high yields of 85% and 62% for the NmCP1 unit. Not only does the synthesis of the bipolar CzCb1 require an additional step, but also the consumption of expensive carboline is high. Finally, the silyl ether protection group was removed and the resulting alcohol monomer precursor was esterified to produce the methacrylates **M5** and **M6**.

### Polymer synthesis

The TADF emitter methacrylate monomer **M1** was synthesized in a one-step procedure as described previously.<sup>26</sup> All copoly-

mers were obtained in a free radical polymerization employing 2,2-azobis(2-methylpropionitrile) (AIBN) as the thermal initiator in 1,4-dioxane at 65 °C (Scheme 3). The molecular compositions and weight distributions of the resulting copolymers **P1**–**7** are presented in Table 1. To ensure the comparability of the copolymers and in line with common device fabrication practices, the TADF emitter (**M1**) content in all copolymers was set to 10 wt%, which corresponds to 6.4 mol% in the carbazole/carboline copolymers **P1**–**3** and 12.7 mol% in the mCP derivative copolymers **P4**–**7**.

Even though the <sup>1</sup>H-NMR spectra of the purified copolymers (Fig. S17–S23†) clearly confirm the presence of the characteristic signals of both the TADF emitter and the respective host species, the polymer composition cannot be determined *via* integration due to the structural similarity of the monomers and the resulting overlap of the signals in the spectrum. Instead, the polymerizations were conducted to high conversions, generally higher than 90% and the integrals of the residual monomer peaks were compared to the initial monomer feed ratio before the polymerization was started. As shown by the exemplarily in Fig. S24,† the ratio of the comonomers before and after the polymerization is almost identical, thus a statistical monomer incorporation in the polymer chain is assumed and the polymer composition can be considered to



**Scheme 3** Free radical polymerizations initiated thermally by AIBN to obtain the carbazole/carboline emitter copolymers **P1–3** and the uni- and bipolar mCP derivative emitter copolymers **P4–7**.

**Table 1** Molecular composition according to monomer feed ratios of copolymers with the TADF emitter and respective host and the molecular weight distributions after size exclusion chromatography in THF (PMMA calibration)

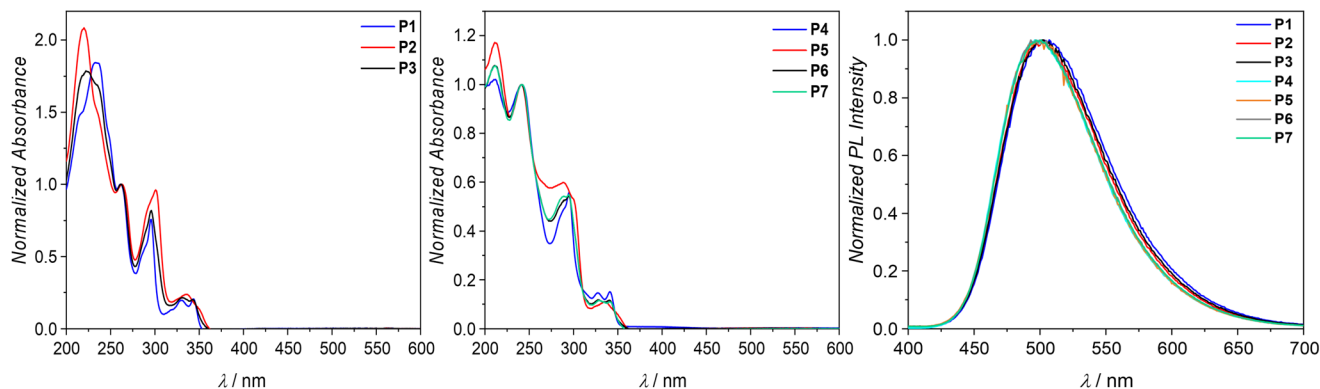
No.	mol% <b>M1</b>	mol% <b>M2</b>	mol% <b>M3</b>	mol% <b>M4</b>	mol% <b>M5</b>	mol% <b>M6</b>	M <sub>n</sub> [kg mol <sup>-1</sup> ]	<i>D</i>
<b>P1</b>	6.4	93.6	—	—	—	—	38	3.1
<b>P2</b>	6.4	—	93.6	—	—	—	33	3.6
<b>P3</b>	6.4	46.8	46.8	—	—	—	30	3.6
<b>P4</b>	12.7	—	—	87.3	—	—	55	2.6
<b>P5</b>	12.7	—	—	—	—	87.3	44	2.9
<b>P6</b>	12.7	—	—	43.65	—	43.65	53	3.0
<b>P7</b>	12.7	—	—	—	87.3	—	45	2.5

match the monomer feed ratio. Furthermore, the molecular weight distribution of the obtained copolymers was determined by size exclusion chromatography in THF (Fig. S25 and S26†).

In addition, the thermal properties of the polymers **P2–6** were studied in a Thermal Gravimetric Analysis (TGA) and a Differential Scanning Calorimetry (DSC) measurement (Fig. S27 and S28†). The polymers **P2** and **P3** based on carbazole and  $\alpha$ -carboline showed a glass transition temperature of 130 and 138 °C respectively, which is in the same temperature range as the previously reported  $T_g$  of **P1** of 137 °C.<sup>22</sup> For the polymers **P4–6** based on derivatives of mCP as internal hosts the detected glass transition temperatures were significantly lower with values around 107 °C despite the larger aromatic systems in the polymer side chains. The reduction of the  $T_g$  of **P4–6** compared to **P1–3** could be attributed to the longer aliphatic linker chains in the host comonomers. However, the thermal stability of the mCP based polymers **P4–6** is superior to **P1–3** with a significant mass loss starting after 320 °C compared to 230 °C in **P2–3**. The overall high thermal stability of all polymers is advantageous for the fabrication process.

To gain more insight into the optoelectronic properties of the synthesised copolymers, the UV-Vis and the photoluminescence spectra were measured of the polymer films **P1–7**. The UV-Vis spectra of the carbazole and carboline polymers **P1** and **P2** are relatively similar with a high optical gap at around 350 nm (Fig. 1, left). As expected, the UV-Vis spectrum of **P3** resembles both. Overall, the UV-Vis-spectra of the mCP derivatives **P4–7** differ only marginally from **P1–3**. Interestingly, the copolymer with the two unipolar monomers **P6** and the copolymer with the bipolar host **P7** produced almost indistinguishable spectra thus confirming the validity of replacing the synthetically challenging bipolar host by a combination of unipolar co-hosts.

The photoluminescence spectra of the polymer films **P1–7** are shown in Fig. 1 (right). While the carbazole copolymer **P1** shows a peak emission at 505 nm, the carboline equivalent **P2** is slightly blue-shifted ( $\lambda_{\max} = 502$  nm). The emission of the carbazole and carboline copolymer **P3**, however, lies between **P1** and **P2**. It appears that the carboline content in the copolymer blue-shifts the emission whereas the (partial) addition of carbazole leads to more de-excitation processes and thus a



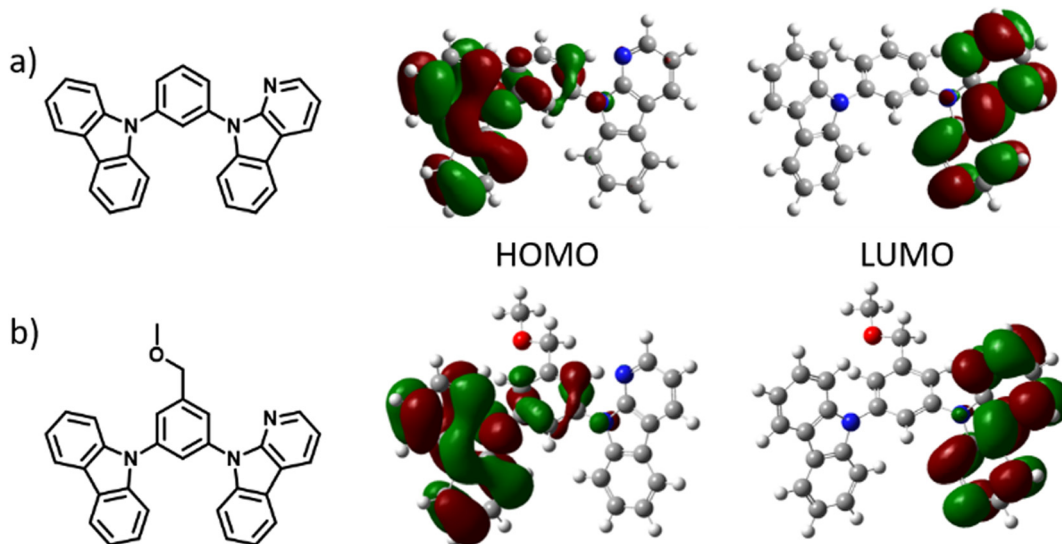
**Fig. 1** UV-Vis spectra of films of copolymers **P1–3** (left) and **P4–7** (middle) and respective photoluminescence spectra with excitation at 290 nm (right).

slight red-shift of the photoluminescence. It is noteworthy that all uni- and bipolar mCP derivative polymers **P4–7** show almost identical emission compared to **P1–3** where a blue-shifted maximum intensity at 498 nm is observed. Judging solely from the photoluminescence spectra, the mCP derivatives in **P4–7** can be considered more suitable hosts than the small molecules carbazole and carboline in **P1–3** alone, even though the energy loss and the associated red shift of the emission is only marginal.

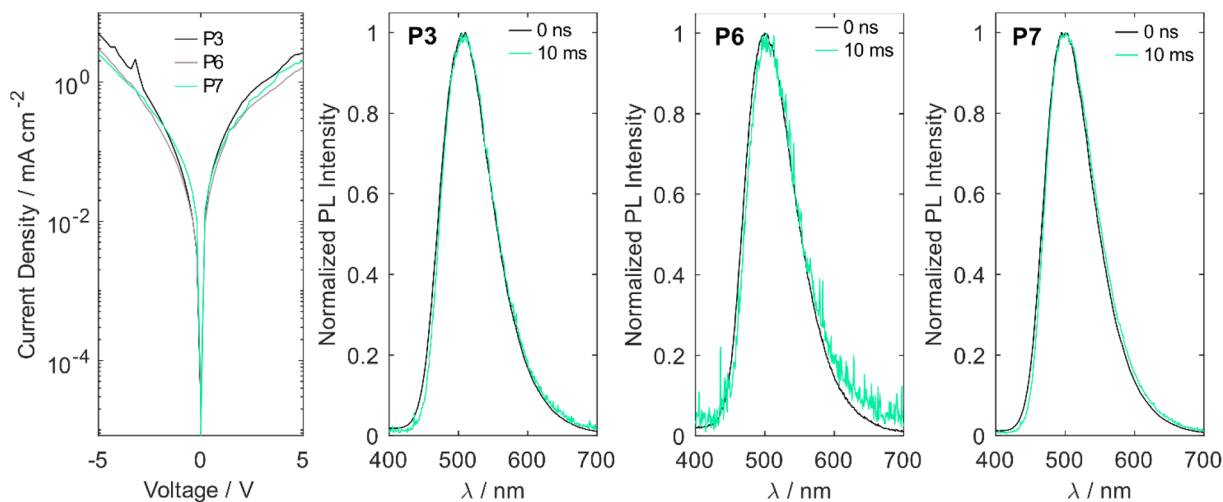
Photoluminescence quantum yields (PLQYs) were measured for all (am)bi-polar hosts materials (**P3**, **P6**, **P7**, Table S1†). **P3** has the lowest PLQY of  $24.9 \pm 1.6\%$ , while the two mCP derivative polymers have higher PLQYs. **P6** had the highest PLQY of  $44.0 \pm 2.8\%$ , and **P7** had a PLQY of  $38.2 \pm 2.5\%$ . This trend is also seen in the single-triplet energy gap ( $\Delta E_{ST}$ ) that was estimated from the onset of PLs of the fluorescence (@ 0 ns) and phosphorescence (@ 10 ms) measured at 77 K (Fig. 3). **P3**, **P6** and **P7** had  $\Delta E_{ST}$ s of 0.43, 0.25 and 0.25 eV, respectively (singlet and triplet energies are displayed in Table S2†). These

energy values are typical for TADF materials and agree with previously reported values for polymers using the same emitting species.<sup>23</sup> To further confirm that the TADF properties of **M1** have been retained, time resolved PL (TRPL) measurements were conducted at room temperature (300 K). The TRPL of **P3**, **P6** and **P7** (Fig. S29†) all indicate that there is both a prompt and delayed lifetime component, where the delayed component is visible in the  $\mu$ s timescale.

In addition, the influence of the linker modification on the frontier orbitals of the aromatic system was investigated to determine if the polymer differs from the small molecule properties. Taking the example of the bipolar small molecule CzCb1 (Fig. 2a), the HOMO and LUMO energy levels were calculated by density functional theory (DFT) and compared to the alkyl linker modified molecule as a model for **M5** (Fig. 2b). The distributions of the frontier orbitals are identical in both molecules, with the HOMO being concentrated on the carbazole and phenyl ring whereas the LUMO is exclusively located on the carboline part of the aromatic system. The calculated



**Fig. 2** DFT calculated distributions of the HOMO and LUMO in the bipolar host CzCb1 (a) and the modified CzCb1 with an alkyl linker (b).



**Fig. 3** Representative current density of hole-only single carrier devices for **P3**, **P6** and **P7** (extreme left) and normalized photoluminescence (PL) spectra of **P3**, **P6** and **P7** (left to right) at 77 K (measured at 0 ns and 10 ms).

energy levels delivered similar values for both molecules (HOMO: CzCb1  $-5.37$  eV vs. CzCb1 with linker  $-5.29$  eV and LUMO: CzCb1  $-1.17$  eV vs. CzCb1 with linker  $-1.15$  eV), which is in line with findings in our previous study, in which cyclic voltammetry measurements of a homopolymer of **M4** delivered HOMO and LUMO energies that did not notably differ from those of the small molecule.<sup>22</sup> It can thus be safely assumed that the energy levels of the synthesized polymers correspond to the values of the small molecules reported in literature without significant deviation (Table S2†). Major structural tuning or conjugation enhancement can lead to significant changes in the HOMO–LUMO values especially in conjugated polymers, but in our case, the polymer backbone is non-conjugated and variations are very minor so dramatic energy level tuning is not expected.

While the polymer of **M1** is well characterized, there are no literature reports about polymers based on the  $\alpha$ -carboline methacrylate **M2**. *Via* cyclic voltammetry of **P2** and the optical gap, the ionization potential and the associated HOMO and LUMO energies were determined as  $-5.43$  eV and  $-1.99$  eV, respectively, which is higher than the reported energy levels of the carbazole polymer (HOMO:  $-5.61$  eV, LUMO:  $-2.10$  eV).<sup>27</sup>

Furthermore, the charge mobility in the polymers was assessed in hole only single charge carrier devices (Fig. 3 and Table S3†). The transport of positive charges in **P7** with the bipolar host species was hereby compared to the copolymers containing a combination of unipolar co-hosts **P6** with the mCP derivatives and **P3** which employs a mixture of carbazole and carboline as pendant groups.

Holes were injected from either electrode (ITO/PEDOT:PSS, or Au/Ag) and the measured current densities were almost identical from both electrodes indicating that the current density measured was most likely a fair representation of only the charge mobility within the materials (**P3**, **P6** or **P7**). As the energy levels of the electrode were deep (PEDOT:PSS  $\sim 5.2$  eV and Au  $\sim 5.1$  eV), electron injection into the device was pre-

vented, which was further supported by the device not “turning on” as an OLED would. Whilst only **P7** carried the bipolar CzCb1 as a pendant group and **P3** and **P6** consist of copolymerized unipolar hosts, it is evident that the hole mobility for all polymers was approximately the same as all of the devices performed within the standard error of each other. Minimal variations between the polymers are attributed to fluctuations in the film thickness of the polymer layer which was determined to be around 20 nm for each of the fabricated single charge carrier devices.

## Conclusions

In summary, a series of non-conjugated TADF copolymers with carbazole,  $\alpha$ -carboline as well as uni- and bipolar derivatives of the host mCP as a pendant group were synthesized. To avoid the synthetically challenging asymmetric bipolar monomer CzCb1, the corresponding unipolar hole- and electron-transporting versions were copolymerized in an equal ratio. Polymers using only carbazole and  $\alpha$ -carboline as host pendant groups showed slightly red-shifted photoluminescence and a lower PLQY than the copolymers based on derivatives of the commercial host mCP. However, among the latter, the combination of the unipolar co-hosts exhibited identical optoelectronic properties as the polymer carrying the bipolar host species, which was further confirmed by PLQY measurements and a single charge carrier device study. Moreover, DFT calculations demonstrated that the modification of the small molecule hosts did not impair the energy levels of the frontier orbitals significantly. Unlike in bipolar host small molecules, the presented strategy allows for a simple adjustment of hole and electron transporting components through variation of the monomer ratio. A more-in-depth study of the performance in ink-jet printed OLEDs of the TADF copolymer **P6** with two unipolar co-hosts is reported elsewhere.<sup>28</sup>

## Conflicts of interest

There are no conflicts to declare.

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