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AUTHOR(S):
Kusakabe, Yu; Wada, Yoshimasa; Misono, Tomoya; Suzuki, Katsuaki; Shizu, Katsuyuki; Kaji, Hironori

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Imidazole Acceptor for Both Vacuum-Processable and Solution-Processable Efficient Blue Thermally Activated Delayed Fluorescence

Yu Kusakabe, Yoshimasa Wada, Tomoya Misono, Katsuaki Suzuki, Katsuyuki Shizu, and Hironori Kaji*

ABSTRACT: The members of the imidazole family have been widely used for electron transporting, host, conventional fluorescent, and phosphorescent materials. Although the imidazole core also has great potential as an acceptor segment of deep-blue thermally activated delayed fluorescence (TADF) owing to its high triplet energy, the emission color of imidazole-based TADF organic light-emitting diodes (OLEDs) has so far been limited to blue to green. In this work, four acridan-imidazole systems are theoretically designed aiming for deep- or pure-blue emitters. All four emitters exhibit deep-blue to blue emission owing to the high energy levels of the lowest excited singlet states, exhibiting y coordinates of Commission Internationale de l’Eclairage coordinates between 0.06 and 0.26. The molecule composed of a trifluoromethyl-substituted benzimidazole acceptor in combination with a tetramethyl-9,10-dihydroacridine donor (named MAc-FBI) achieves a high maximum external quantum efficiency (EQE\text{MAX}) of 13.7% in its application to vacuum-processed OLEDs. The emitter has high solubility even in ecofriendly nonhalogenated solvents, which motivates us to fabricate solution-processed MAc-FBI-based OLEDs, resulting in an even higher EQE\text{MAX} of 16.1%.

1. INTRODUCTION

To boost the performance of the organic light-emitting diodes (OLEDs), thermally activated delayed fluorescence (TADF) materials have been widely adopted to OLEDs as emitters.\textsuperscript{1–8} This is because TADF materials can convert all singlet and triplet excitons into light through reverse intersystem crossing (RISC) owing to the small energy difference ($\Delta$E\text{ST}) between the lowest excited singlet state ($S_1$) and the lowest triplet state ($T_1$). To achieve small $\Delta$E\text{ST}, the separation between the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) is valid. Therefore, the combination of electron donor and acceptor fragments has been incorporated into TADF molecular design. Imidazole is a widely used molecular fragment for OLED materials such as electron transporting materials, host materials, and emitter materials. This is because of their good tunability of photophysical and electrical properties derived from both an electron-donating and an electron-accepting imidazole core, as has been reviewed in ref 9. Thus, imidazole is one of the good candidates for the molecular fragments of TADF materials. So far, five reports have been found for TADF emitters that possess an imidazole core in their application to OLEDs.\textsuperscript{10–14} Wang and co-workers reported two phenanthroimidazole-based green TADF emitters, named PPZTPI and PPZPPI.\textsuperscript{10} For benzimidazole (BI)-based TADF emitters, Adachi and co-workers reported a green emitter named DHPZ-2BI,\textsuperscript{11} and Zysman-Colman and co-workers also reported four green emitters.\textsuperscript{12} Kido and co-workers reported a series of BI-based emitters and one of which was named PXZ-BIP exhibited sky-blue emission in Commission Internationale de l’Eclairage (CIE) coordinates ($x, y$) of (0.22, 0.42).\textsuperscript{13} Lu and co-workers reported dicyanoimidazole-based emitters named imM-SPAC and imM-DMAC that showed blue emissions with CIE coordinates of (0.17, 0.18) and (0.16, 0.19), respectively.\textsuperscript{14}

Considering the high LUMO energy level and high $T_1$ energy of the imidazole core, we can expect efficient deep- or pure-blue TADF materials by appropriate molecular modification. In this work, we investigate four donor–acceptor-type emitters (Figure 1) using BI-derivatives as acceptors in combination with 9,10-dihydro-9,9-dimethylacridine (Ac) electron-donating derivatives. All emitters exhibited blue emission ranging from deep-blue to blue in their doped films. Among the four, MAc-FBI (Figure 1), which has the combination of a 2,7,9,9-tetramethyl-9,10-dihydroacridine donor (MAc) and 1-phenyl-2-(2-( trifluoromethyl)phenyl)-

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1H-benzo[d]imidazole (FBI), exhibits efficient TADF and achieves a EQE_{MAX} = 13.7% with blue emission of CIE (0.15, 0.19) in its application to vacuum-processed OLEDs. In addition, MAc-FBI is found to be highly soluble in non-

Table 1. TD-DFT Calculated and Experimental Data of Ac-BI, MAc-BI, Ac-FBI, and MAc-FBI

| emitter   | HOMO [eV] | LUMO [eV] | E_g [eV] | S_1 [eV] | T_1 [eV] | T_2 [eV] | T_3 [eV] | ΔE_{ST} [eV] |
|-----------|----------|-----------|----------|----------|----------|----------|----------|-------------|
| Ac-BI     | −5.46    | −1.38     | 4.09     | 3.30^a/3.44^b | 3.12^a/3.00^b | 3.13     | 3.28     | 0.18^a/0.44^b |
| MAc-BI    | −5.25    | −1.34     | 3.90     | 3.13^a/3.28^b | 3.06^a/2.95^b | 3.12     | 3.14     | 0.07^a/0.33^b |
| Ac-FBI    | −5.61    | −1.66     | 3.95     | 3.10^a/3.19^b | 3.06^a/2.92^b | 3.15     | 3.22     | 0.04^a/0.27^b |
| MAc-FBI   | −5.39    | −1.63     | 3.76     | 2.92^a/3.08^b | 2.89^a/2.94^b | 3.07     | 3.23     | 0.03^a/0.14^b |

^aCalculated values. ^bExperimental values.
halogenated solvents, which motivated us to fabricate solution-processed OLEDs as well. The EQE_{MAX} reached 16.1% with blue emission of CIE (0.17, 0.24). Both vacuum- and solution-processable features of MAc-FBI make it an attractive TADF emitter for OLEDs.

2. RESULTS AND DISCUSSION

2.1. Molecular Design. The four molecules, Ac-BI, MAc-BI, Ac-FBI, and MAc-FBI, were designed using Ac as an electron donor core and BI as an acceptor core (Figure 1). Here, calculations were conducted using Gaussian 16,13 and the results are summarized in Table 1, Figures 1, and S5. Molecular geometries were optimized using density functional theory (DFT) at the ground state (S0) with the PBE0/6-311+ +G(d,p) level of theory considering toluene solvent polarity effects using the polarizable continuum model (PCM). Calculations for the excited states were also conducted using time-dependent DFT (TD-DFT) with the same basis set and the PCM condition using the S0 optimized structure. Torsion angles between donors and acceptors were large (89–90°), which resulted in adequate separation of the HOMO and LUMO distributed on the donor and acceptor units, respectively (Figure S1). The HOMO energy level of MAc-BI and MAc-FBI was higher than that of Ac-BI and Ac-FBI because of CH3 substitution in Ac. Also, owing to the CF3 substitution in BI, the LUMO energy level of Ac-FBI and MAc-FBI was lower than that of Ac-BI and MAc-BI, as we designed. The calculated ΔE_ST values for the S0 structures were within 0.2 eV for the four molecules; Ac-BI (0.18 eV), MAc-BI (0.07 eV), Ac-FBI (0.04 eV), and MAc-FBI (0.03 eV), as shown in Table 1.

The origin of the difference in ΔE_ST between the four emitters was investigated by natural transition orbital (NTO) analysis. The highest occupied NTO (HONTO) and lowest unoccupied NTO (LUNTO) are provided in Figure 1. For all of the molecules, the HONTOs and LUNTOs of S1 were distributed on the donors and acceptors, respectively, which is characteristic of charge transfer (CT) states. The energy gaps between the CT-type S1 and the lowest CT-type or CT-dominant hybridized local CT (HLCT)-type triplet (T1 for Ac-BI, T2 for MAc-BI, T3 for Ac-FBI, and T1 for MAc-FBI) were small. In contrast to HLCT T2, T3 for Ac-FBI and MAc-FBI, the T1s of Ac-BI and MAc-BI were locally excited type triplet (3LE) states. Owing to the lower-lying 3LE in energy, these two molecules, especially Ac-BI, exhibited relatively large ΔE_ST. From HOMO energy levels, LUMO energy levels, and NTO analysis, thanks to CH3 and CF3 substitution in Ac and BI, respectively, appropriate CT-type S1 energies were expected for the small ΔE_ST in MAc-BI, Ac-FBI, and especially MAc-FBI.

2.2. Photophysical Property of Emitters. The four molecules were synthesized and purified by train sublimation for further photophysical measurements (see the Supporting Information for details). Figure 2a exhibits their UV−vis absorption and photoluminescence (PL) spectra in a 10−5 M toluene solution. A broad and weak absorption shoulder with an absorption coefficient ε of ~2000 M−1 cm−1 was observed at approximately 350–400 nm. The absorption energies decreased in the order of Ac-BI, MAc-BI, Ac-FBI, and MAc-FBI. All of the molecules exhibited blue emission. Emission spectra at room temperature were red-shifted in the order of a maximum emission wavelength (λ_{MAX}) of 422 nm (Ac-BI), 442 nm (MAc-BI), 463 nm (Ac-FBI), and 485 nm (MAc-FBI). The origin of the di...
The photophysical performances of the four emitters in the solid films were investigated using spin-coated amorphous films prepared from chloroform solution. Emitters were doped at a concentration of 10 wt% in a host matrix, bis[2-(diphenylphosphino)phenyl]ether oxide (DPEPO) or 9-(4-tert-butylphenyl)-3,6-bis(triphenylsilyl)-9H-carbazole (CzSi), which are typically used for blue TADF emitters. As summarized in Table 2, Ac-BI exhibited the deepest blue emission among the four molecules, with CIE (0.16, 0.20) in DPEPO and (0.16, 0.11) in CzSi. Even the most red-shifted emission among the four molecules, with CIE (0.16, 0.08) in 3), to obtain the delayed fluorescence from T1, respectively.

Table 2. Photophysical Properties of Ac-BI, MAc-BI, Ac-FBI, and MAc-FBI in a DPEPO and CzSi Host Spin-coated Film at an Excitation Wavelength of 300 nm

| host   | emitter | λ_{max} [nm] | CIE (x, y) | Φ_{PL} [%] | k_{nr} (S_{1→S_{0}}) [10^6 s^{-1}] | k_{p} (T_{1→S_{0}}) [10^7 s^{-1}] | k_{ISC} [10^7 s^{-1}] | k_{RISC} [10^8 s^{-1}] |
|--------|---------|--------------|-------------|------------|-------------------------------------|---------------------------------|------------------------|-----------------------|
| DPEPO  | Ac-BI   | 416^{a}/419^{a} | (0.16, 0.07) | 20.7 ± 0.1 | Detected                            |                                 |                        |                       |
|        | MAc-BI  | 440^{a}/440^{a} | (0.16, 0.12) | 31.6 ± 0.7 |                                     |                                 |                        |                       |
|        | Ac-FBI  | 462^{a}/451^{a} | (0.16, 0.15) | 56.2 ± 0.1 |                                     |                                 |                        |                       |
|        | MAc-FBI | 482^{a}/473^{a} | (0.18, 0.26) | 64.2 ± 0.4 |                                     |                                 |                        |                       |
| CzSi   | Ac-BI   | 410^{a}/412^{a} | (0.17, 0.06) | 18.9 ± 0.1 |                                     |                                 |                        |                       |
|        | Ac-FBI  | 426^{a}/429^{a} | (0.16, 0.08) | 20.3 ± 0.0 |                                     |                                 |                        |                       |
|        | MAc-FBI | 439^{a}/446^{a} | (0.16, 0.11) | 29.2 ± 0.1 |                                     |                                 |                        |                       |

aΦ_{PL} were determined to be average value ± error (95% confidence interval). Measured under an air condition. N2 flow.

Table 3. Values of τ_{p}, τ_{d}, Φ_{PL}, k_{nr}, k_{ISC}, and k_{RISC} of 10 wt% Emitter-Doped Films, Where Emitters Were Ac-BI, Ac-FBI, and MAc-FBI

| host   | emitter | τ_{p} [ns] | τ_{d} [μs] | Φ_{PL} [%] | k_{nr} (S_{1→S_{0}}) [10^6 s^{-1}] | k_{ISC} [10^7 s^{-1}] | k_{RISC} [10^8 s^{-1}] |
|--------|---------|------------|-----------|------------|-------------------------------------|------------------------|-----------------------|
| DPEPO  | Ac-BI   | 5.7        | 22.2      | 8.8        | 15.3                                | 6.2                    | 15.9                  | 2.4                   |
|        | Ac-FBI  | 12.8       | 69.7      | 10.9       | 45.3                                | 8.6                    | 7.1                   | 7.0                   |
|        | MAc-FBI | 22.8       | 33.3      | 13.2       | 51.0                                | 5.8                    | 12.4                  | 3.8                   |
| CzSi   | Ac-BI   | 10.8       | 86.2      | 14.1       | 15.1                                | 13.1                   | 9.6                   | 8.0                   |
|        | Ac-FBI  | 14.9       | 66.6      | 8.9        | 38.0                                | 6.0                    | 8.8                   | 6.1                   |

aΦ_{PL}, k_{nr}, and k_{ISC} are the Φ_{PL} of the prompt component, Φ_{PL} of the delayed component, the rate constant of radiative decay from S_{1}, and the rate constant of nonradiative decay from T_{1}, respectively. Rate constants were deduced based on the previously reported method.

Figure 3. Photographs of 10 wt% emitter: CzSi spin-coated film. From left to right, Ac-BI, MAc-BI, Ac-FBI, and MAc-FBI.

The collected fluorescence and phosphorescence spectra at 77 K using DPEPO as the host (Figure S6). The S_{1}, T_{1}, and ΔE_{ST} values of all emitters in toluene solutions and in DPEPO films are summarized in Table S3.

Photoluminescence quantum yields (PLQYs) and transient PL decay curves are shown in Table 2 (also see Figure S7). Ac-BI exhibited a low PLQY of ~20% and did not show clear delayed fluorescence in both DPEPO and CzSi. MAc-BI exhibited almost the same PLQY of 20%, and no clear delayed emission was observed in CzSi, while a higher PLQY of 30% was obtained, and delayed fluorescence was observed in DPEPO. Ac-FBI displayed delayed fluorescence in both CzSi and DPEPO. The PLQY was still low (29%) in CzSi, while an improved high PLQY of 56% was obtained in DPEPO. MAc-FBI exhibited the best performance: clear delayed fluorescence in both hosts and relatively high PLQYs of 47% in CzSi and 56% in DPEPO. From temperature-dependent transient PL measurement for MAc-BI, Ac-FBI, and MAc-FBI doped in DPEPO films, the delayed fluorescence was confirmed to be thermal activated (see Figure S8). These TADF characteristics and PLQYs are well explained by the above-mentioned experimental ΔE_{ST} of the four emitters.

Table 3 shows the prompt lifetime (τ_{p}) and delayed lifetime (τ_{d}) of the materials detected delayed fluorescence, determined from the fitting of the PL decay curves with double exponential functions. The τ_{d} were relatively long (20–90 μs). To obtain deeper insight into the long τ_{d} we determined rate constants for radiative (k_{p}), nonradiative (k_{nr}), ISC (k_{ISC}), and RISC (k_{RISC}) transitions based on the previously reported method (Table 3). MAc-BI, Ac-FBI, and MAc-FBI exhibited k_{RISC} of 10^{5–7} s^{-1}, leading to delayed PL lifetimes on the order of 20–90 μs. Owing to the greater k_{ISC} compared to k_{p} and k_{nr}, photoexcited singlet excitons tend to be converted into triplet excitons. Then, some excitons are assumed to decay nonradiatively, resulting in moderate PLQYs, as shown in Table 2.

2.3. Organic Light-Emitting Diodes. We fabricated both vacuum- and solution-processed OLEDs using the MAc-FBI emitter doped in the CzSi host to achieve efficient blue emission. As shown in Figure S9a, the device structure of the vacuum-processed OLED was indium–tin–oxide (ITO) (50 nm)/4,4′-cyclohexyldiindenecis[N,N-bis(4-methylphenyl)benzeneamine] (TAPC) (60 nm)/1,3-bis(9,9-dimethylacridin-10(9H)-yl)benzene (mAP) (10 nm)/10 vol % MAc-BI:CzSi (20 nm)/2,8-bis(diphenylphosphoryl) dibenzol[b,d]furan (PPF) (10 nm)/1,3-bis[3,5-di(pyridin-3-yl)phenyl]benzene (BmPyPhB) (30 nm)/lithium quinolin-8-olate (LiQ) (1 nm)/Al (80 nm). As shown in Figure 4 and Table S4, the OLED exhibited blue emission with CIE (0.15, 0.19) and EQE_{MAX} was 13.7%. We also investigated the solution-processed OLEDs with a device structure of ITO (50 nm)/poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEO) (80 nm)/MAc-FBI (30 nm)/lithium quinolin-8-olate (LiQ) (1 nm)/Al (80 nm). As shown in Figure S9b, the device structure of the solution-processed OLED was indium–tin–oxide (ITO) (50 nm)/poly(3,4-ethylenedioxythiophene) polystyrene sulfonate (PEO) (80 nm)/MAc-FBI (30 nm)/lithium quinolin-8-olate (LiQ) (1 nm)/Al (80 nm). As shown in Figure 4 and Table S4, the OLED exhibited blue emission with CIE (0.15, 0.19) and EQE_{MAX} was 13.7%.
DOT:PSS) (35 nm)/poly(9-vinylcarbazole) (PVK) (10 nm)/
10 wt % MAC-FBI:CzSi (20 nm)/PPF (5 nm)/1,3,5-tris(1-
phenyl-IH-benzo[d]imidazol-2-yl)benzene (TPBi) (50 nm)/
LiF (1 nm)/Al (80 nm) (see Figure S9b). Here, the
PEDOT:PSS, PVK, and 10 wt % MAC-FBI:CzSi layers were
fabricated by spin-coating and the following layers were
vacuum deposited. We prepared an emitter layer solution using
toluene because MAC-FBI is highly soluble in toluene with a
solubility of more than 10 mg mL\(^{-1}\). The high solubility in
such a nonhalogenated solvent is a great advantage for the
fabrication of solution-processed multilayer OLEDs\(^{22}\) and the
halogen-free process is attractive for environmentally friendly
OLED production.\(^{23,24}\) Compared with the vacuum-deposited OLED, the solution-processed OLED exhibited a higher
EQE\(_{\text{MAX}}\) of 16.1% with blue emission of CIE (0.17, 0.24) (Figure S3 and Table S4). These good
device performances of MAc-FBI in both vacuum-processed
and solution-processed OLEDs demonstrate that the imidazole
core is a promising candidate for designing blue TADF.

3. CONCLUSIONS

We report four emitters, Ac-BI, MAc-BI, Ac-FBI, and MAc-
FBI, using an acridan-benzimidazole donor–acceptor system.
From the theoretical calculations, the four emitters exhibited a
sufficiently high \(S_1\) energy to show blue emission, owing to the
wide HOMO–LUMO gap, which results from the combina-
tion of the deep HOMO of the Ac core and the shallow
LUMO of the BI core. In addition to the high \(S_1\), MAc-FBI
exhibited the smallest \(\Delta E_{\text{ST}}\) among the four emitters, owing
to the good combination of MAC and FBI, which were modified
from Ac and BI, respectively. Experimentally, all four molecules
exhibited blue emissions. MAC-BI, Ac-FBI, and MAC-FBI
exhibited the typical photophysical feature of TADF. Vacuum-
processed OLEDs using an MAC-FBI emitter exhibited an
EQE\(_{\text{MAX}}\) of 13.7% with blue emission of CIE (0.15, 0.19).
Furthermore, MAC-FBI was found to be highly soluble in an
environment-friendly nonhalogenated solvent. Solution-pro-
cessed OLEDs using MAC-FBI as an emitter were fabricated
and exhibited a higher EQE\(_{\text{MAX}}\) of 16.1% with blue emission of
CIE (0.17, 0.24). Both the vacuum- and solution-processability
of MAC-FBI demonstrated in this work reveals part of the
potential of an imidazole core for the design of deep- or pure-
blue TADF materials.

**ASSOCIATED CONTENT**

**Supporting Information**

The Supporting Information is available free of charge at https://pubs.acs.org/doi/10.1021/acsomega.2c01308.
Experimental procedure including synthesis, photophysical measurements, preparation of the samples, and device characterization and additional scheme or figure or table of synthesis, theoretical investigation, photophysical properties, and device performances (PDF)

**AUTHOR INFORMATION**

**Corresponding Author**
Hironori Kaji – Institute for Chemical Research, Kyoto University, Uji, Kyoto 611-0011, Japan; orcid.org/0000-0002-5111-3852; Email: kaji@scl.kyoto-u.ac.jp

**Authors**
Yu Kusakabe – Institute for Chemical Research, Kyoto University, Uji, Kyoto 611-0011, Japan
Yoshimasa Wada – Institute for Chemical Research, Kyoto University, Uji, Kyoto 611-0011, Japan; orcid.org/0000-0001-6139-8794
Tomoya Misono – Institute for Chemical Research, Kyoto University, Uji, Kyoto 611-0011, Japan
Katsuki Suzuki – Institute for Chemical Research, Kyoto University, Uji, Kyoto 611-0011, Japan
Katsuyuki Shizu – Institute for Chemical Research, Kyoto University, Uji, Kyoto 611-0011, Japan; orcid.org/0000-0002-1835-0418

Complete contact information is available at: https://pubs.acs.org/10.1021/acsomega.2c01308

**Author Contributions**
Y.K., Y.W., T.M., and K.S. conducted quantum chemical calculations; T.M. and K.S. contributed to the synthesis of the materials; T.M. performed material characterization; Y.K. contributed to the device structure; Y.K., Y.W., and T.M. contributed to device fabrication; and H.K. planned and supervised the study. All authors wrote the manuscript and approved the final version of the manuscript.

**Notes**
The authors declare no competing financial interest.

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