Organic Electroluminescent Materials Realizing Efficient Conversion from Electricity to Light

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Thermally activated delayed fluorescence (TADF) emitters are attracting increasing attention because of their high electricity-to-light conversion efficiencies in organic light-emitting diodes (OLEDs). The electricity-to-light conversion efficiency of TADF emitters depends largely on their rates of radiative decay and transition from triplet to excited singlet states. To realize high-performance TADF-based OLEDs, TADF emitters should exhibit both rapid radiative decay and rapid triplet-to-singlet conversion. Herein, we describe a rational molecular design strategy for such TADF emitters based on theoretical and computational chemistry. Our design strategy provides a guideline for realizing high-performance TADF-based OLEDs.

Keywords: organic light-emitting diode, thermally activated delayed fluorescence, transition dipole moment, vibronic coupling, overlap density

1. Introduction

Organic light-emitting diodes [1,2] (OLEDs) are a promising technology for next-generation solid-state lighting sources and flexible full-color flat-panel displays. OLEDs have the advantages of greater design flexibility and being lighter in weight than conventional lighting devices. In OLEDs, holes and electrons are injected from an anode and cathode, respectively, and recombine to generate singlet and triplet excitons in the light-emitting layer. Excitons generated from electron-hole pairs are divided into singlet and triplet excitons in a ratio of 1:3 according to spin statistics. Consequently, singlet and triplet excitons account for 25% and 75% of the total electrogenerated excitons, respectively.

In the early development of OLEDs, normal fluorescent emitters were used as emissive dopants. In OLEDs containing normal fluorescent emitters (called fluorescent OLEDs herein), only singlet excitons are converted into light, while triplet excitons are lost as heat. Hence, the efficiency with which electron-hole pairs are converted into light, called the internal quantum efficiency (IQE), is limited to 25% (Figure 1a).

The use of phosphorescent materials as emitting dopants enables more effective conversion of triplet excitons into light [3]. Phosphorescent emitters can also convert singlet excitons into light by phosphorescence following intersystem crossing (ISC) from their excited singlet states to triplet states. Hence, the IQE of phosphorescent OLEDs is potentially 100% (Figure 1b), which means...
that all of the electrogenerared excitons can be converted into light. In 2001, an IQE of nearly 100% was achieved using an iridium complex as an emissive dopant [4].

Thermally activated delayed fluorescence (TADF) emitters are a promising alternative to phosphorescent emitters in OLEDs [5] as they can convert all of the electrogenerared excitons into light as fluorescence. TADF-based OLEDs have been reported with IQEs approaching 100% and their device performance is now competitive with that of phosphorescent OLEDs [6,7]. TADF emitters convert triplet excitons into light as delayed fluorescence via reverse ISC (RISC) from their triplet to excited singlet states (Figure 1c). Efficient TADF requires a small energy difference ($\Delta E_{ST}$, Figure 2) between the excited singlet and triplet states because the RISC is driven by thermal activation.

Figure 2 shows radiative and non-radiative transitions involved in a TADF process. For simplicity, only the ground state ($S_0$), the lowest excited singlet state ($S_1$), and the lowest triplet state ($T_1$) are included. $\Delta E_{ST}$ is the energy difference between $S_1$ and $T_1$. Generally, TADF emitters show both normal fluorescence ($S_1 \rightarrow S_0$) and delayed fluorescence ($T_1 \rightarrow S_1 \rightarrow S_0$). The lifetime of delayed fluorescence (µs–ms) is much longer than that of normal fluorescence (ns) because delayed fluorescence involves spin flipping in the $T_1 \rightarrow S_1$ RISC process. In contrast, the emission spectra of normal and delayed fluorescence are generally identical. This is because both normal and delayed fluorescence are radiative decays from the same excited singlet state, $S_1$.

TADF-based OLEDs show an efficiency roll-off caused by triplet-triplet annihilation (TTA) and singlet-triplet annihilation (STA) [8] resulting from bimolecular $T_1-T_1$ and $S_1-T_1$ interactions, respectively. Hence, suppressing bimolecular interactions by rapidly converting $T_1$ and $S_1$ into light and shortening their lifetimes can improve the efficiency roll-off. A short $T_1$ lifetime can be realized by increasing the transition dipole moment $\mu_{10}$ between $S_1$ and $S_0$ and thereby increasing the rate constant of the $S_1 \rightarrow T_0$ radiative decay ($k_r$). By increasing $\mu_{10}$, $k_r$ can be made greater than the rate constants of $S_1 \rightarrow T_1$ ISC and $S_1 \rightarrow S_0$ non-radiative decay ($k_{ISC}$ and $k_{nr}$, respectively), leading to quantitative conversion from $S_1$ to $S_0$.

If a small $\Delta E_{ST}$ and large $\mu_{10}$ are compatible, both singlet and triplet excitons generated in TADF-based OLEDs are rapidly converted into light and their efficiency roll-offs are expected to be improved. However, there is generally a trade-off between having a small $\Delta E_{ST}$ and a large $\mu_{10}$. When the electronic configuration of $S_1$ is written as a one-electron excitation from an occupied molecular orbital ($\phi_{occ}$) to an unoccupied molecular orbital ($\phi_{unocc}$), $\Delta E_{ST}$ can be expressed approximately by

$$\Delta E_{ST} \approx 2e \int_{\mathbb{R}^3} \frac{e^{2}(\phi_{occ}(\mathbf{r}_1)\phi_{unocc}(\mathbf{r}_2))\phi_{occ}(\mathbf{r}_2)\phi_{unocc}(\mathbf{r}_1)}{||\mathbf{r}_1 - \mathbf{r}_2||} d\mathbf{r}_1 d\mathbf{r}_2, \quad (1)$$

where $e$ is the elementary charge and $\mathbf{r}_1$ and $\mathbf{r}_2$ represent the positions of electrons 1 and 2, respectively, in three-dimensional space. Eq. 1 shows that $\Delta E_{ST}$ is small when $\phi_{occ}(\mathbf{r}_1)\phi_{unocc}(\mathbf{r}_2)$ and $\phi_{occ}(\mathbf{r}_2)\phi_{unocc}(\mathbf{r}_1)$ are small and this is realized by reducing the
DACT-II shows a strong performance in 3,6-diamine (DACT-II, Figure 3) [6].

Δ have developed TADF emitters with various configuration of $S_1$, $\Phi_1$, and is the dominant contributor to the electronic wave functions of $S_1$. $\Phi_1$ and $\Phi_0$. When the one-electron excitation from $\varphi_{\text{occ}}$ to $\varphi_{\text{unocc}}$ is the key mechanism for TADF. Eqs. 2 and 3 show that $\mu_{10}$ is large when the spatial overlap between $\varphi_{\text{occ}}$ and $\varphi_{\text{unocc}}$ is large, especially in regions distant from the coordinate origin. This can be realized by increasing the spatial overlap between $\varphi_{\text{occ}}$ and $\varphi_{\text{unocc}}$. Thus, a trade-off exists between having a small $\Delta E_{ST}$ and a large $\mu_{10}$.

We have tried to optimize this trade-off and have developed TADF emitters with various $\Delta E_{ST}$ and $\mu_{10}$ values [6,9,10]. In 2012, we reported a TADF emitter with a twisted donor-acceptor structure, 2-[4-(phenoxazin-10-yl)phenyl]-4,6-diphenyl-1,3,5-triazine (PXZ-TRZ, Figure 3) [9]. PXZ-TRZ has a small $\Delta E_{ST}$ of 0.07 eV [11] and shows efficient TADF. Using PXZ-TRZ as an emitting dopant, we prepared efficient TADF-based OLEDs with an external quantum efficiency (EQE) of 12%. To improve the luminescence efficiency of PXZ-TRZ, we chemically modified its electron-donating unit to develop another efficient TADF emitter, $N^1$-[4-(4,6-diphenyl-1,3,5-triazin-2-yl)phenyl]-$N^2$-[4-(diphenylamino)-phenyl]-$N^3$-diphenylbenzene-1,4-diamine (DPA-TRZ, Figure 3) [10]. DPA-TRZ has a large $\mu_{10}$, leading to a photoluminescence quantum yield (PLQY) of nearly 100%. However, the $\Delta E_{ST}$ of DPA-TRZ is much larger than that of PXZ-TRZ. By further optimizing the electron-donating unit, we have solved the trade-off between $\Delta E_{ST}$ and $\mu_{10}$ to obtain a highly efficient TADF emitter, 9-[4-(4,6-diphenyl-1,3,5-triazin-2-yl)phenyl]-$N,N,N',N''$-tetraphenyl-9H-carbazole-3,6-diamine (DACT-II, Figure 3) [6]. DACT-II shows a strong performance in OLEDs with a high EQE and low efficiency roll-off because of its small $\Delta E_{ST}$ and large $\mu_{10}$.

Here, we describe in detail the molecular design strategy for TADF emitters to have a small $\Delta E_{ST}$ and a large $\mu_{10}$, which is based on density functional theory (DFT) and time-dependent DFT (TD-DFT) methods. By comparing calculated $\varphi_{\text{occ}}$, $\varphi_{\text{unocc}}$, $\rho_{10}$, $\mu_{10}$, and $\Delta E_{ST}$ of PXZ-TRZ, DPA-TRZ, and DACT-II, we illustrate how the trade-off between $\Delta E_{ST}$ and $\mu_{10}$ has been solved.

2. Method of calculating $\varphi_{\text{occ}}$, $\varphi_{\text{unocc}}$, $\rho_{10}$, $\mu_{10}$, and $\Delta E_{ST}$

The geometry optimization of $S_0$ for isolated PXZ-TRZ, DPA-TRZ, and DACT-II molecules were carried out using the PBE0 functional and the correlation-consistent polarized valence double-zeta (cc-pVDZ) basis set as implemented in the Gaussian 09 program package [12-14]. The computational level used in the present manuscript is different from those used in the previous papers [6,9,10]. Using the optimized $S_0$ geometries, six low-lying excited singlet states ($S_1$–$S_6$) and six low-lying excited triplet states ($T_1$–$T_6$) were calculated using the time-dependent DFT method as implemented in the Gaussian 09 program package. $\Delta E_{ST}$ was then calculated by subtracting the $S_0$-$T_1$ excitation energy from the $S_0$-$S_1$ excitation energy. $\rho_{10}$ and $\mu_{10}$ were calculated using $\Phi_0$ and $\Phi_1$ obtained from the DFT and TD-DFT methods. Electronic configurations with configuration interaction coefficients larger than $1.0 \times 10^{-4}$ were used in the calculations of $\rho_{10}$ and $\mu_{10}$.

3. Molecular design of TADF emitters with a small $\Delta E_{ST}$

For molecules containing a donor-acceptor structure, $\varphi_{\text{occ}}$ and $\varphi_{\text{unocc}}$ are predominantly
localized on the electron-donating and electron-accepting units, respectively. Hence, \( \varphi_{\text{occ}} \) and \( \varphi_{\text{unocc}} \) tend to be well separated, making donor-acceptor-type molecules promising candidates for TADF emitters. The donor-acceptor emitter PXZ-TRZ was designed to have a small \( \Delta E_{\text{ST}} \). PXZ-TRZ shows efficient TADF and has a PLQY of 66% in a solid-state host matrix [9]. PXZ-TRZ contains a phenoxazine donor unit and a triphenyltriazine acceptor unit (Figure 3). The \( \varphi_{\text{occ}} \) and \( \varphi_{\text{unocc}} \) of PXZ-TRZ are the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO), respectively. The steric repulsion between hydrogen atoms near the linkage position induces a large torsion angle of (88.9°) between the electron-donating and accepting units. This highly twisted structure provides PXZ-TRZ with well-separated HOMO and LUMO distributions, with the HOMO predominantly localized on the electron-donating unit, while the LUMO is predominantly localized on the electron-accepting unit (Figure 4a). This orbital distribution leads to the small experimentally determined \( \Delta E_{\text{ST}} \) of 0.07 eV and efficient \( T_1 \rightarrow S_1 \) RISC. However, the highly twisted structure results in the undesirable effects of reducing \( \mu_{10} \) (Table 1), suppressing \( S_1 \rightarrow S_0 \) radiative decay, and consequently lowering the PLQY. Figure 4a shows the \( \rho_{10} \) distribution of PXZ-TRZ. The sparse distribution of \( \rho_{10} \) is reflected in the small \( \mu_{10} \) of PXZ-TRZ.

Table 1. \( \mu_{10}, \sum_m(V_{10}^{m})^2, \) and \( \Delta E_{\text{ST}} \) for PXZ-TRZ, DPA-TRZ, and DACT-II calculated at the PBE0/cc-pVDZ level of theory. The functional and basis set used in this work are different from those reported previously (see ref 6, 9, and 10).

| Compound   | \( \mu_{10} \) (a.u.) | \( \sum_m(V_{10}^{m})^2 \) (a.u.) | \( \Delta E_{\text{ST}} \) (eV) |
|------------|------------------------|------------------------------------|-----------------------------|
| PXZ-TRZ    | 3.17×10^{-4}           | 3.12×10^{-8}                      | 0.009                       |
| DPA-TRZ    | 2.98                   | 5.57×10^{-7}                      | 0.482                       |
| DACT-II    | 1.87                   | 2.28×10^{-7}                      | 0.155                       |

4. Molecular design of TADF emitters with a large \( \mu_{10} \)

DPA-TRZ was designed to have enhanced \( S_1 \rightarrow S_0 \) radiative decay and a higher PLQY than PXZ-TRZ. DPA-TRZ contains three electron-donating diphenylamino units and the same triphenyltriazine acceptor unit. The HOMO→LUMO+1 excitation and HOMO→LUMO excitation of DPA-TRZ were assigned as \( S_1 \) and \( S_2 \), respectively. The \( S_1 \) (HOMO→LUMO+1 excitation) was chosen as the luminescent state because the transition dipole moment of \( S_1 \) (2.98 a.u.) is much larger than that of \( S_2 \) (0.04 a.u.), that is, \( \varphi_{\text{occ}} \) and \( \varphi_{\text{unocc}} \) are the HOMO and LUMO+1, respectively. The \( S_1 \) and \( S_2 \) levels of DPA-TRZ are nearly degenerate in energy (2.83 and 2.87 eV for \( S_1 \) and \( S_2 \), respectively) and this is because the LUMO and LUMO+1 of triphenyltriazine are nearly degenerate [15]. The relative order of the HOMO→LUMO+1 and HOMO→LUMO excitation energies depended on the choice of functionals and basis sets. When the M06-2X/cc-pVDZ level of theory was used, the HOMO→LUMO excitation was the luminescent state and assigned as \( S_1 \) [10].

The torsion angle between the electron-donating and accepting units of DPA-TRZ (29.5°) is smaller than that of PXZ-TRZ (89.9°) because the steric repulsion between the hydrogen atoms near the linkage position in DPA-TRZ is weaker than that in...
PXZ-TRZ. Hence, the HOMO and LUMO are more widely distributed across DPA-TRZ than in PXZ-TRZ (Figure 4a, b). Consequently, $\rho_{10}$ is much denser in DPA-TRZ than in PXZ-TRZ (Figure 4a, b), leading to the larger $\mu_{10}$ of DPA-TRZ (2.98 a.u., Table 1) than that of PXZ-TRZ (3.17×10^{-4} a.u., Table 1). Although the increase in the HOMO-LUMO spatial overlap resulted in the undesirable effect of increasing $\Delta E_{ST}$, the experimental $\Delta E_{ST}$ of DPA-TRZ (0.11 eV) is sufficiently small to allow efficient $T_1\rightarrow S_1$ RISC and thereby TADF. DPA-TRZ therefore shows efficient TADF and has a PLQY of nearly 100% when doped into a host matrix [10]. The outermost diphenylamino groups play a role in withdrawing the HOMO distribution away from the electron-accepting unit, which limits excessive HOMO-LUMO spatial overlap that would cause a large $\Delta E_{ST}$. The calculated $\Delta E_{ST}$s and their relative orders of magnitude were helpful in designing TADF emitters but they were found to be in poor agreement with experimentally obtained values.

5. Molecular design of TADF emitters with a small $\Delta E_{ST}$ and a large $\mu_{10}$

As described above, careful molecular design is required to realize efficient TADF emitters because of the trade-off between having a small $\Delta E_{ST}$ and a large $\mu_{10}$. DACT-II was obtained by replacing the central diarylaminato unit of DPA-TRZ with a carbazole unit. The electron-donating unit of DACT-II [3,6-bis(diphenylamino)carbazole, DAC] is a promising building block for realizing large $\mu_{10}$ and has been used in various efficient TADF emitters [16,17]. The $\varphi_{occ}$ and $\varphi_{unocc}$ of DACT-II are the HOMO and the LUMO, respectively. The distributions of the HOMO and LUMO are more localized in DACT-II than in DPA-TRZ (Figure 4b, c). Consequently, the experimentally determined $\Delta E_{ST}$ is significantly reduced to 0.009 eV [6], which is smaller than $k_BT$ at room temperature (0.026 eV). This suggests that $T_1\rightarrow S_1$ RISC induced by thermal activation would readily occur at room temperature. Importantly, the $\mu_{10}$ of DACT-II is also relatively large, although it is smaller than that of DPA-TRZ. DACT-II has a smaller $\mu_{10}$ than DPA-TRZ because the $\rho_{10}$ distribution of DACT-II is less dense than that of DPA-TRZ (Figure 4b, c). Thus, by carefully controlling $\rho_{10}$, we can solve the trade-off between having a small $\Delta E_{ST}$ and a large $\mu_{10}$.

Its small $\Delta E_{ST}$ and relatively large $\mu_{10}$ provide DACT-II with both rapid $S_1\rightarrow S_0$ radiative decay and rapid $T_1\rightarrow S_1$ RISC [6], indicating that DACT-II can rapidly convert both electrogenerated singlet and triplet excitons into light. DACT-II shows efficient TADF and has a PLQY of nearly 100% [6]. OLEDs containing DACT-II as the emissive dopant showed high EQEs up to 29.6%, which is one of the highest EQEs reported for TADF-based OLEDs. The corresponding IQE was nearly 100% and furthermore this high IQE was maintained at high luminance levels [6].

6. Influence of the chemical modification of donor units on $S_1\rightarrow S_0$ non-radiative decay

$S_1\rightarrow S_0$ non-radiative decay competes with $S_1\rightarrow S_0$ radiative decay and can lower the luminescence efficiency of TADF emitter molecules. Hence, the influence of chemical modification on $S_1\rightarrow S_0$ non-radiative decay should be considered when designing TADF emitters. $S_1\rightarrow S_0$ non-radiative decay is caused by intramolecular vibronic coupling, the strength of which is measured by the vibronic coupling constant, $V_{10}^{m}$ [10] given by

$$V_{10}^{m} = \int \rho_{nm}(x) \left[ -\sum_{\nu} \frac{Z_{\nu} \mathbf{e}_{\nu}^{(m)} \cdot \mathbf{R}_{\nu}}{\sqrt{M_{\nu}}} \right] dx,$$

where the index $m$ represents the $m^{th}$ vibrational mode and $Z_{\nu}$, $M_{\nu}$, and $\mathbf{R}_{\nu}$ are the charge, mass, and position of the $A^{th}$ nucleus, respectively, and $\mathbf{e}_{\nu}^{(m)}$ is a three-dimensional vector representing the relative displacement of the $A^{th}$ nucleus for the $m^{th}$ vibrational mode. From Eq. 4, $V_{10}^{m}$ increases with increasing $\rho_{10}$. The sum of squared $V_{10}^{m}$, $\Sigma_m(V_{10}^{m})^2$, is a rough measure of $k_{nr}$. Generally, molecules having higher $V_{10}^{m}$’s have consequently fast $k_{nr}$ and small $V_{10}^{m}$ values are therefore desirable for TADF emitter molecules. The calculated $\Sigma_m(V_{10}^{m})^2$ values for PXZ-TRZ, DPA-TRZ, and DACT-II show that PXZ-TRZ is expected to have the slowest $k_{nr}$. However, its $\mu_{10}$ is quite small and radiative and non-radiative decay compete, which results in the lowest PLQY of 66%. Although the $\Sigma_m(V_{10}^{m})^2$ values are larger for DPA-TRZ and DACT-II, their $\mu_{10}$s are sufficiently large that
non-radiative decay pathways are suppressed and consequently their PLQYs are nearly 100%.

7. Conclusion
TADF emitters with a small $\Delta E_{ST}$ and a large $\mu_{10}$ are expected to show rapid $S_1\rightarrow S_0$ radiative decay and $T_1\rightarrow S_1$ RISC and hence are promising emitting dopants for high performance OLEDs. We described in detail the molecular design strategy for realizing TADF emitters with a small $\Delta E_{ST}$ and a large $\mu_{10}$. By carefully controlling the $\rho_{10}$ distribution, it is possible to achieve a small $\Delta E_{ST}$ and a large $\mu_{10}$. Our molecular design strategy is useful for developing TADF emitters, leading to high-performance OLEDs with low efficiency roll-off.

Acknowledgements
This work was supported by JSPS KAKENHI Grant Number 15K17900. The computations were partly carried out using the computer facilities at the Academic Center for Computing and Media Studies (ACCMS), Kyoto University and the SuperComputer System, Institute for Chemical Research, Kyoto University.

References
1. C. W. Tang and S. A. VanSlyke, Appl. Phys. Lett., 51 (1987) 913.
2. C. W. Tang, S. A. VanSlyke and C. H. Chen, J. Appl. Phys., 65 (1989) 3610.
3. M. A. Baldo, D. F. O’Brien, Y. You, A. Shoustikov, S. Sibley, M. E. Thompson and S. R. Forrest, Nature, 395 (1998) 151.
4. C. Adachi, M. A. Baldo, M. E. Thompson and S. R. Forrest, J. Appl. Phys., 90 (2001) 5048.
5. H. Uoyama, K. Goushi, K. Shizu, H. Nomura and C. Adachi, Nature, 492 (2012) 234.
6. H. Kaji, H. Suzuki, T. Fukushima, K. Shizu, K. Suzuki, S. Kubo, T. Kominoh, H. Oiwa, F. Suzuki, A. Wakamiya, Y. Murata and C. Adachi, Nat. Commun., 6 (2015) 8476.
7. J. W. Sun, J.-H. Lee, C.-K. Moon, K.-H. Kim, H. Shin and J.-J. Kim, Adv. Mater., 26 (2014) 5684.
8. K. Masui, H. Nakano and C. Adachi, Org. Electron., 14 (2013) 2721.
9. H. Tanaka, K. Shizu, H. Miyazaki and C. Adachi, Chem. Commun., 48 (2012) 11392.
10. K. Shizu, M. Uejima, H. Nomura, T. Sato, K. Tanaka, H. Kaji and C. Adachi, Phys. Rev. Applied, 3 (2015) 014001.
11. H. Nakano, T. Higuchi, T. Furukawa, K. Masui, K. Morimoto, M. Numata, H. Tanaka, Y. Sagara, T. Yasuda and C. Adachi, Nat. Commun., 5 (2014) 4016.
12. C. Adamo and V. Barone, J. Chem. Phys., 110 (1999) 6158.
13. J. T. H. Dunning, J. Chem. Phys., 90 (1989) 1007.
14. M. J. Frisch, G. W. Trucks, H. B. Schlegel, G. E. Scuseria, M. A. Robb, J. R. Cheeseman, G. Scalmani, V. Barone, B. Mennucci, G. A. Petersson, H. Nakatsuji, M. Caricato, X. Li, H. P. Hratchian, A. F. Izmaylov, J. Bloino, G. Zheng, J. L. Sonnenberg, M. Hada, M. Ehara, K. Toyota, R. Fukuda, J. Hasegawa, M. Ishida, T. Nakajima, Y. Honda, O. Kitao, H. Nakai, T. Vreven, J. A. Montgomery, J. E. Peralta, F. Ogliaro, M. Bearpark, J. J. Heyd, E. Brothers, K. N. Kudin, V. N. Staroverov, R. Kobayashi, J. Normand, K. Raghavachari, A. Rendell, J. C. Burant, S. S. Iyengar, J. Tomasi, M. Cossi, N. Rega, J. M. Millam, M. Klene, J. E. Knox, J. B. Cross, V. Bakken, C. Adamo, J. Jaramillo, R. Gomperts, R. E. Stratmann, O. Yazyev, A. J. Austin, R. Cammi, C. Pomelli, J. W. Ochterski, R. L. Martin, K. Morokuma, V. G. Zakrzewski, G. A. Voth, P. Salvador, J. J. Dannenberg, S. Dapprich, A. D. Daniels, Ö. Farkas, J. B. Foresman, J. V. Ortiz, J. Cioslowski and D. J. Fox, Gaussian 09, Revision C.01. GAUSSIAN 09, Revision C.01, Gaussian, Inc., Wallingford, CT, 2009.
15. K. Shizu, Y. Sakai, H. Tanaka, S. Hirata, C. Adachi and H. Kaji, ITE Trans. on MTA, 3 (2015) 108.
16. K. Shizu, J. Lee, H. Tanaka, H. Nomura, T. Yasuda, H. Kaji and C. Adachi, Pure Appl. Chem., 87 (2015) 627.
17. K. Shizu, H. Tanaka, M. Uejima, T. Sato, K. Tanaka, H. Kaji and C. Adachi, J. Phys. Chem. C, 119 (2015) 1291.