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# Solvent dependent ultrafast dynamics of multi-branched thiophene-based triphenylamine derivatives with a triazine core

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# ABSTRACT

In the present study, we investigated the solvent effect on ultrafast dynamics of the intramolecular charge transfer (ICT) state in multi-branched thiophene-based triphenylamine derivatives with a triazine core (STAPA-a and STAPA-b). The experimental results showed that the fluorescence of STAPA-a and STAPA-b originated from the ICT state. In a weakly polar solvent toluene, the relaxation of the ICT state showed mono-exponential decay due to the absence of the influence of solvent polarity, which directly reflected the fluorescence lifetime. In polar solvents tetrahydrofuran and dichloromethane, accelerated evolutions of ICT states and multi-exponential processes were exhibited because of the interaction between solvent and solute. Furthermore, to explore the influence of the molecular structure on the dynamics of the ICT state, we compared the fluorescence relaxation processes in solvents with different viscosity. We found that the time-resolved properties of STAPA-b exhibit significant differences due to the introduction of alkyl chains.

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## 1. Introduction

In recent years, triphenylamine (TPA) derivatives have attracted wide attention due to their excellent optical properties. They have exhibited great application potential in organic light emitting diodes [1–4], solar cells [5–8], and photorefractive [9] and nonlinear optical materials [10–12]. This excellent optical performance originates from the strong electron donating ability of TPA. As the electron donor, the molecule with TPA can easily constitute D- $\pi$ -D and D- $\pi$ -A structures, which have remarkable intramolecular charge transfer (ICT) features. More and more TPA derivatives have been synthesized. However, the majority have focused on developing the synthetic method of these derivatives, with limited understanding of the ICT processes involved.

Generally, the ICT process occurs between acceptor and donor units. Therefore, revealing the mechanisms of fluorescence evolution caused by ICT is of great significance to the materials containing donor and acceptor units [13–15]. It is well known

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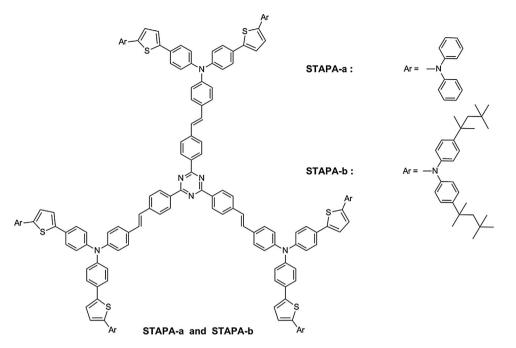


Fig. 1. Molecular structures of STAPA based compounds: STAPA-a and STAPA-b.

that the polarity of materials will change during the ICT process, especially in a solution. Both the polarity and viscosity of the solvent can influence the ICT process, recognized as the solvent effect. Thus, investigation into the solvent effect can provide useful information of ICT mechanisms [16–22].

In our previous work, we synthesized the multi-branched TPA that showed good nonlinear optical properties [23,24] due to their strong electron donating ability [25,26]. To thoroughly investigate these types of materials, we further synthesized two octupolar triazine molecules (STAPA-a and STAPA-b) containing multi-branched thiophene-based TPA derivatives with and without alkyl groups. They exhibited excellent nonlinear optical ability resulting from the charge transfer process [27]. In the present study, we focused on exploring the ICT process in these materials. We studied the ICT properties of the two new derivatives (STAPA-a and STAPA-b) by steady state and time-resolved optical spectroscopy in different solvents with various polarity and viscosity. We found that the fluorescence decay processes exhibited mono-exponential decay in a weakly polar solvent and triple-exponential decay in polar solvents. The polarity of the solvent can accelerate the formation and relaxation of ICT states. Furthermore, we found that the solvent viscosity also significantly influenced the relaxation processes, due to the distortion degree affected by the introduction of the alkyl chain in STAPA-b.

## 2. Experimental

#### 2.1. Materials

The compounds STAPA-a and STAPA-b, whose molecular structures are illustrated in Fig. 1, were synthesized from condensation of aldehyde (with or without long octyl chains) with triazine moiety by the Horner-Wadsworth-Emmons reaction. Detailed information of synthesis and characterization of STAPA-a and STAPA-b can be found in a previously published paper [27]. In these two compounds, TPA served as an electron donor, 1, 3, 5-triazine as an electron acceptor, and styryl as a  $\pi$ bridge. The introduction of a thiophene moiety with rich  $\pi$ -electron density enriches the  $\pi$ -electron density of the system and improves the optoelectronic properties. Octupolar molecules consisting of a strong triazine electron-accepting center and an electron-donating end group linked via a  $\pi$ -conjugated bridge are excellent optical materials.

## 2.2. Experimental setup

The steady-state absorption spectra were recorded by a UV–vis spectrophotometer (TU-1901, Purkinje General). The fluorescence properties were measured by a spectrometer (iHR550, HORIBA) combined with a time-correlated single photon counting system (TCSPC, PicoHarp 300). In this setup, under excitation by a femtosecond pulse laser, simultaneous recording of the steady state and transient fluorescence spectra of samples can occur by switching the collected fluorescence to different detectors (charge-coupled device or single photon avalanche diodes). In the TCSPC configuration, tuning the spectrometer enabled the measurement of the fluorescence decay profile at various detection wavelengths. The response time of the instrument was approximately 40 ps.

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