

# Synthesis and Photophysical Properties of Two Intramolecular Charge-transfer Chromophores obtained by [2+2] Cycloaddition of TCNE and TCNQ to an Electron-rich Alkyne

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*A disubstituted alkyne containing triphenylamine and phenyl groups as key compound, was prepared by Sonogashira cross-coupling reaction between phenylacetylene and (4-iodophenyl)diphenylamine. Cycloaddition of two strong electron acceptors, i.e., tetracyanoethylene (TCNE) and 7,7,8,8-tetracyanoquinodimethane (TCNQ) to this electron-rich alkyne led to compounds (TPA-CN and TPA-CNQ) containing both donor and acceptor groups. All compounds were characterized by <sup>1</sup>H-NMR and FTIR spectroscopy, while the photophysical and electrochemical properties were investigated by UV-Vis, fluorescence spectroscopy and cyclic voltammetry.*

*Keywords: Electron rich triphenylamine derivative, electron acceptors, tetracyanoethylene, 7,7,8,8-tetracyanoquinodimethane, photophysical properties, intramolecular charge transfer interactions.*

Conjugated polymers have attracted much interest from both a scientific point of view and for their potential applications in micro- and nano-electronics and optoelectronics [1]. Their application in organic light-emitting diodes [2-7] and organic field effect transistors [8,9] has been reported. The latest attempts for their use as active elements in solar energy conversion are very encouraging and much effort has recently been directed to this field [10-12].

Triphenylamine (TPA) derivatives are an important class of compounds for the realization of solution processable, low cost and large area electronics [13-15]. TPA as structural unit is responsible for the good photoconducting and hole transporting properties and can be incorporated into a broad range of small molecules (e.g., linear, star-shaped oligomers and dendrimers) and polymers (e.g., homopolymers, copolymers, branched and hyperbranched (co)polymers). These materials have shown excellent solubility, thermal and electrochemical stability, electron donating ability, and optoelectronic properties. They have been used most widely as the hole-transporting layer in electroluminescent devices due to their amorphous film-forming ability and their high carrier mobility. They also can act as efficient dyes for sensitized solar cells.

Since Gratzel et al [16] have reported the first dye-sensitized organic solar cell, a considerable interest for small conjugated molecules having donor and acceptor groups has appeared. These compounds show intramolecular charge-transfer (ICT) interactions and efficient electron transfer from donor to acceptor upon photoexcitation, being promising candidates for building of the dye-sensitized and bulk heterojunction (BHJ) solar cells [11,12]. Diederich et al. [17-19] have recently reported that a variety of electron-rich alkynes have reacted with TCNE to give donor-substituted 1,1,4,4-tetracyano-1,3-butadiene in excellent yields. Furthermore, this reaction was expanded to other strong acceptor molecules, such as 7,7,8,8-tetracyanoquinodimethane (TCNQ) and other donors [20-24].

In this paper, we have synthesized two new intramolecular charge transfer (ICT) compounds by incorporating the triphenylamine moiety (electron donor)

with different cyano groups (electron acceptor) to explore the relationship between chemical structure and properties of these compounds. Properties of the triphenylamine-based derivatives were investigated by spectral methods, such as <sup>1</sup>H-NMR, FT-IR, cyclic voltammetry, UV-Vis and fluorescence spectroscopy.

## Experimental part

### Materials, equipment and methods

Triphenylamine (TPA), phenylacetylene, PdCl<sub>2</sub>·2 PPh<sub>3</sub>, CuI, triphenylphosphine (PPh<sub>3</sub>) (all from Sigma-Aldrich), tetracyanoethylene (TCNE) and 7,7,8,8-tetracyanoquinodimethane (TCNQ) (Fluka) were commercial products. (4-Iodophenyl)diphenylamine was synthesized starting from triphenylamine by iodination with KI/KIO<sub>3</sub> in glacial acetic acid as described in the literature [25]. Solvents such as triethylamine, carbon tetrachloride, methylene chloride and hexane were obtained from commercial sources and used as received or dried by known methods.

The FT-IR spectra were recorded in KBr pellets on a DIGILAB-FTS 2000 spectrometer. UV-Vis and fluorescence measurements were carried out in solution using spectrophotometric grade solvents, on a Specord 200 spectrophotometer and Perkin Elmer LS 55 apparatus, respectively. <sup>1</sup>H-NMR spectra were recorded at room temperature on a Bruker Avance DRX-400 spectrometer (400 MHz) as solutions in CDCl<sub>3</sub>, and chemical shifts are reported in ppm and referenced to TMS as internal standard. The cyclic voltammograms (CV) were recorded using a Bioanalytical System, Potentiostat-Galvanostat (BAS 100B/W) system. The electrochemical cell was equipped with three electrodes: a working electrode (Pt, Φ=1.6 mm), an auxiliary electrode (platinum wire), and a reference electrode (consisted of a silver wire coated with AgCl). Before experiments, working electrode was sonicated in a mixture of detergent and methanol for 5 min and then rinsed with a large amount of doubly distilled water. The reference electrode (Ag/AgCl) was calibrated at the beginning of the experiments by running the CV of ferrocene as the internal standard in an identical cell without any compound in the system. Prior to the each

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experiment, the  $\text{Bu}_4\text{NBF}_4$  solutions were deoxygenated by passing dry argon gas for 10 min. All measurements were performed at room temperature (25°C) under nitrogen atmosphere.

### Synthesis of (4-iodophenyl)diphenylamine (**2**)

A solution of triphenylamine (5g, 20.38 mmol) and KI (2.23 g, 13.45 mmol) in glacial acetic acid (80 mL) was maintained at 85°C for 5h. Then the reaction mixture was cooled at room temperature and  $\text{KIO}_3$  (1.43 g, 6.725 mmol) was added into reaction and additionally stirred at 85°C for 5 h. After cooling the solution was poured into water, the precipitate was filtered and dried. The resulted solid was washed several times with a diluted solution of  $\text{Na}_2\text{S}_2\text{O}_3$ , and finally with water. (4-Iodophenyl) diphenyl-amine (**2**) was obtained after twice recrystallizations from ethyl acetate, to obtain 4.6 g of white-solid, 61.9 %.  $^1\text{H-NMR}$  ( $\text{CDCl}_3$ ): 7.52-7.48 (d, 2H), 7.27-7.21 (m, 4 H), 7.09-6.98 (m, 6H), 6.83-6.80 (d, 2H).

### Synthesis of alkyne arylamine (**3**)

A solution obtained from  $(\text{PPh}_3)_2\text{PdCl}_2$  (0.011g, 0.015 mmol), CuI (0.018 g, 0.094 mmol),  $\text{PPh}_3$  (0.017 g, 0.064 mmol), phenylacetylene (1 mL) and 2 mL triethylamine was introduced in a 50 mL round-bottomed flask. and stirred under dry  $\text{N}_2$  at 50-60°C for 1 hr. Then, a mixture of (4-iodophenyl)diphenylamine (**2**) (1.5 g, 4.04 mmol) and TEA (6.5 mL) was added. After 12 hr of stirring at 60°C, TEA was removed by distillation and solid product was dissolved in ethylic ether, washed with diluted HCl, water and dried over  $\text{MgSO}_4$ . The product after purification is obtained as viscous oil that crystallized in time.

FT-IR ( $\text{cm}^{-1}$ , KBr): 3435, 3033-2853 ( $\nu\text{C-H}$ ), 2202 ( $-\text{Ca}\equiv\text{C}-$ ), 1588 ( $\text{C}=\text{C}$ , conjugated phenyl groups), 1488, 1316, 1277 ( $-\text{CN}$  stretching vibration), 1070, 1025, 831, 754, 689, 497.

$^1\text{H-NMR}$  ( $\text{CDCl}_3$ ): 7.52-7.48 (m, 4H), 7.41-7.20 (m, 9 H), 7.12-6.97 (m, 6H).

### Synthesis of compounds TPA-CN and TPA-CNQ

To a solution of compound **3** (0.43 g, 1.24 mmol) in  $\text{CCl}_4$  (15 mL) was added TCNE (0.23 g, 1.86 mmol) or TCNQ (0.379g, 1.86 mmol). The mixture was stirred at reflux for 24 h in argon atmosphere. The solutions were filtered, and after evaporation of the solvent, the solids were dissolved in a minimal amount of  $\text{CH}_2\text{Cl}_2$ , then *n*-hexane was added to precipitate the final product. Compound TPA-CN was

isolated in 63.7% yield as dark red solid. In the case of compound TPA-CNQ, pure product was obtained in 35% yield as dark green glassy solid.

TPA-CN: Mp 156-157°C; FT-IR ( $\text{cm}^{-1}$ , KBr): 3433, 2923-2853 ( $\nu\text{C-H}$ ), 2220 ( $-\text{Ca}\equiv\text{N}-$ ), 1587 ( $\text{C}=\text{C}$ , conjugated phenyl groups), 1487, 1336, 1204, 757, 697.  $^1\text{H-NMR}$  ( $\text{CDCl}_3$ ): 7.73-7.71 (d, 2H), 7.66-7.62 (t, 3H), 7.57-7.52 (d, 2H), 7.42-7.38 (m, 4H), 7.28-7.21 (m, 6H), 6.94-6.92 (d, 2H). MS (ESI,  $m/z$ ): 475.88 ( $\text{M}+\text{H}^+$ ).

TPA-CNQ: Mp 209-210°C; FT-IR ( $\text{cm}^{-1}$ , KBr): 3436, 3051-2923 ( $\nu\text{C-H}$ ), 2222 ( $-\text{Ca}\equiv\text{N}-$ ), 1586 ( $\text{C}=\text{C}$ , conjugated phenyl groups), 1488, 1276, 757, 697, 695.  $^1\text{H-NMR}$  ( $\text{CDCl}_3$ ): 7.54-7.51 (d, 2H), 7.31-7.30 (t, 3H), 7.26-7.21 (m, 6H), 7.08-7.04 (m, 6H), 7.02-6.97 (m, 4H), 6.94-6.92 (dd, 2H). MS (ESI,  $m/z$ ): 549.76 ( $\text{M}+\text{H}^+$ ).

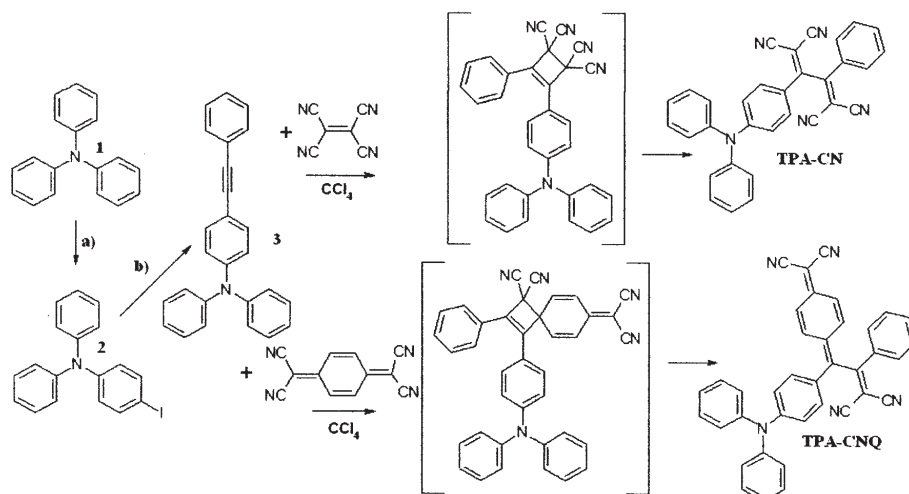
## Results and discussions

### Synthesis

The steps for synthesis of the two donor-acceptor triphenylamine-based oligomers, starting from triphenylamine, are outlined in scheme 1. Iodination of triphenylamine (**1**) with KI/ $\text{KIO}_3$  in AcOH, at 85°C for 5h provided (4-iodophenyl)diphenylamine (**2**). The Sonogashira coupling of **2** with phenylacetylene in high excess and in the presence of catalysts [ $(\text{PPh}_3)_2\text{PdCl}_2$ , CuI,  $\text{PPh}_3$ ] using as solvent triethylamine gave compound **3**. FTIR spectrum of **3** evidences the stretching vibration of triple bond at 2202  $\text{cm}^{-1}$  (fig. 1) while  $^1\text{H-NMR}$  spectrum confirms the structure of the product (see Experimental part). Compound **3**, was treated with tetracyanoethylene (TCNE) to obtain compound TPA-CN or with 7,7,8,8-tetracyanoquinodimethane (TCNQ) to obtain compound TPA-CNQ (scheme 1).

The FT-IR spectra (fig. 1) and  $^1\text{H-NMR}$  spectra in  $\text{CDCl}_3$  (see Supporting Information) have confirmed the expected oligomers structures. FT-IR spectroscopy shows the presence of the characteristic bands of the chromophoric groups in oligomers: the weak vibrational peak at 2202  $\text{cm}^{-1}$  assigned to the internal  $\text{C}\equiv\text{C}$  linkage of compound **3** was replaced by the new absorption peaks emerged at 2220  $\text{cm}^{-1}$  (for TPA-CN) and 2222  $\text{cm}^{-1}$  (for TPA-CNQ), corresponding to the  $\text{Ca}\equiv\text{N}$  stretching vibration. The intensity of  $\text{C}\equiv\text{N}$  peak is higher than of  $\text{C}\equiv\text{C}$  linkage, due to the presence of four CN groups in a molecule.

The  $^1\text{H-NMR}$  spectra of all compounds have proved their structure and proton assignments are presented in figures 2-4.



Scheme 1. Synthetic route for synthesis of compounds TPA-CN and TPA-CNQ  
a)- KI/ $\text{KIO}_3$ , AcOH, 85°C, 5h; b)- [ $(\text{PPh}_3)_2\text{PdCl}_2$ , CuI,  $\text{PPh}_3$ ], TEA, phenylacetylene, 50-60°C, 24 h

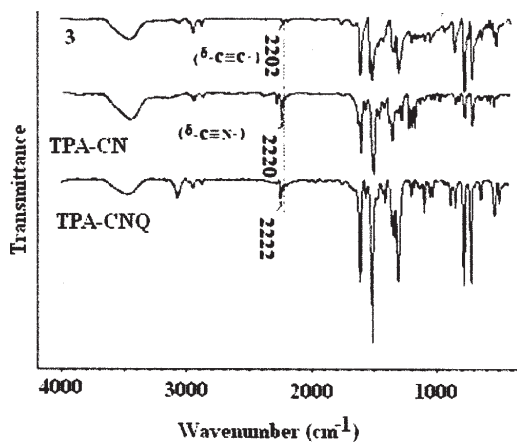


Fig. 1. FTIR spectra of compounds: **3**, TPA-CN and TPA-CNQ

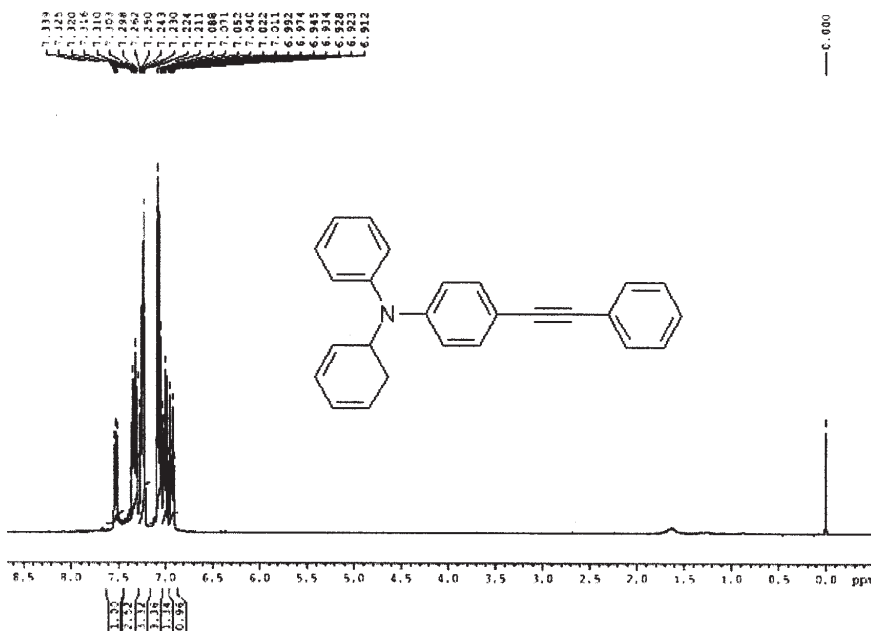


Fig. 2. <sup>1</sup>H-NMR spectrum of **3** (CDCl<sub>3</sub>, 400 MHz)

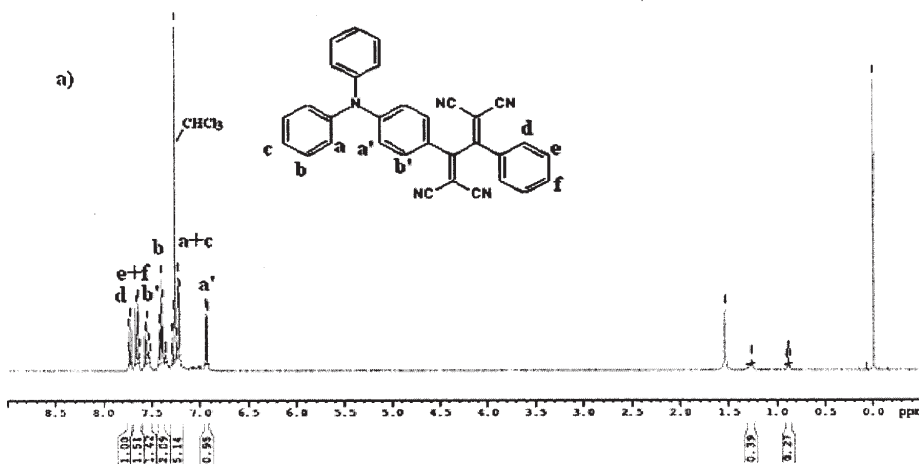


Fig. 3. <sup>1</sup>H-NMR spectra of TPA-CN in CDCl<sub>3</sub> at 20°C. Signals from 0.5-2.5 ppm region are due to impurities from CDCl<sub>3</sub> and synthesis.

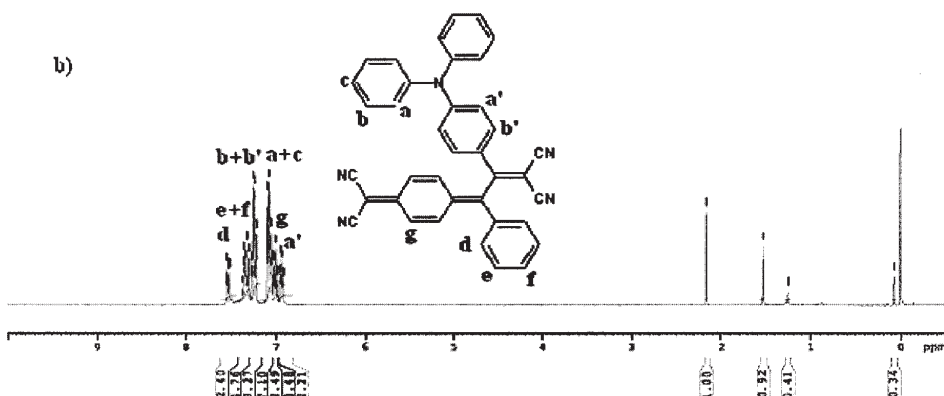


Fig. 4. <sup>1</sup>H-NMR spectra of TPA-CNQ in CDCl<sub>3</sub> at 20°C. Signals from 0.5-2.5 ppm region are due to impurities from CDCl<sub>3</sub> and synthesis

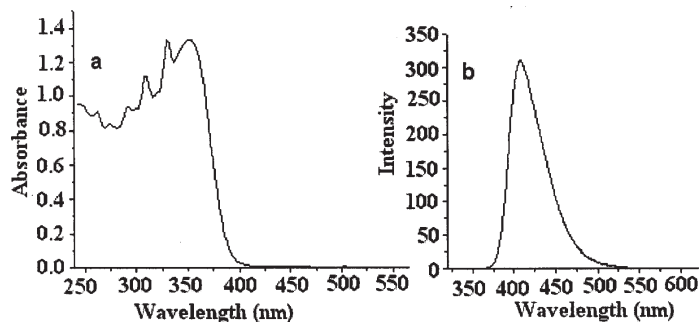


Fig. 5. Absorption (a) and fluorescence (b) spectra of **3** (in  $\text{CHCl}_3$ )

### Optical properties

The UV-Vis spectrum of the parent compound (**3**) displays two distinct absorption regions with maxima at 250-325 nm and 350-400 nm (fig. 5). The first absorption is assigned to  $\pi-\pi^*$  transition in aromatic rings while the second one is due to  $\pi-\pi^*$  transition in whole conjugated molecule. The fluorescence spectrum shows a maximum at 407 nm.

The photophysical properties (the absorption and photoluminescence spectra) of TPA-CN and TPA-CNQ were studied in various polar solvents: chloroform, methylene chloride, dimethylformamide and toluene because they contain both donor and acceptor groups and solvent polarity has a strong influence on them. Figure 6 shows the absorption and photoluminescence (PL) spectra in different solvents while the photophysical data are collected in table 1. The introduction of electron-withdrawing pendant groups by reaction with tetracyanoethylene and 7,7,8,8-tetracyanoquino-dimethane, introduces new absorption bands in UV-Vis spectrum with their maximum shifted from 398 to 850 nm, assignable to intramolecular charge transfer (ICT) interactions between the triphenylamine moiety and the pendant acceptor groups. Depending on the polarity of the solvents, the ICT band is shifted for the two oligomers. Solvatochromism was observed as a characteristic feature of dipolar molecules. The UV-Vis spectrum for the compound TPA-CN, shows a hypsochromic shifting of the ICT bands from 494 nm (chloroform) to 476 nm (dimethylformamide) with

increasing polarity solvents. It is assumed that the first ground state has a larger dipole moment than of the excited-state. Comparison of the data for compound TPA-CNQ, the UV-Vis spectrum displays a bathochromic shift (red) of the ICT bands from 394 nm (toluene) to 850 nm (dimethylformamide) with increasing polarity solvents. In addition, the presence of TCNQ moiety in compound TPA-CNQ, results in a more complex absorption band ( $\lambda_{\text{max}} = 850 \text{ nm}$ ), which is obviously different from compound TPA-CN, due the longest conjugated system. For the emission spectra (PL) in various organic solvents, the oligomers TPA-CN and TPA-CNQ displayed different behaviours. A red-shifting at higher wavelength (bathochromic effect) of the maximum absorption from 389 nm to the 439 nm, for compound TPA-CN was observed, while in the case of compound TPA-CNQ, the photoluminescence spectra exhibited a red-shifted at higher wavelength (bathochromic effect) of the emission maximum at 449 nm for less-polar (toluene) and 465 nm for the most polar solvent (dimethylformamide). For chlorinated solvents ( $\text{CHCl}_3$  and  $\text{CH}_2\text{Cl}_2$ ), the emission spectra displayed emission maxima at 462-463 nm.

The absorption spectra of TPA-CN and TPA-CNQ were measured in various organic solvents and the  $\lambda_{\text{max}}$  values were plotted as a function of solvent polarity  $E_T(30)$  (fig. 7). The oligomers displayed positive solvatochromism, which indicates the greater dipole moment for compound TPA-CNQ and negative solvatochromism for TPA-CN, with increasing of solvent polarity. Finally, it can be observed

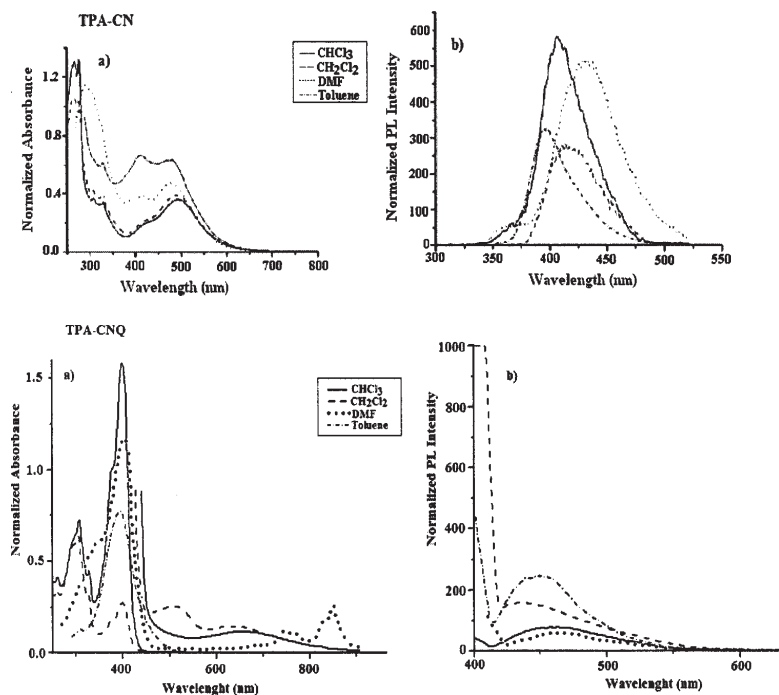


Fig. 6. UV-Vis (a) and photoluminescence (b) spectra of the oligomers in different solvents ( $\text{CH}_2\text{Cl}_2$ ,  $\text{CHCl}_3$ , DMF and toluene)

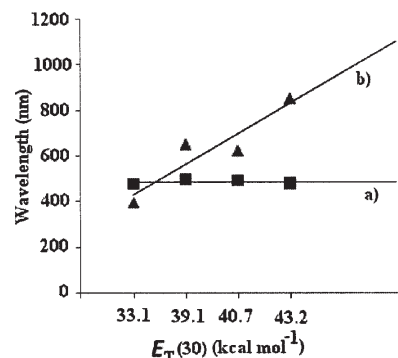


Fig. 7. Relationship between the solvent polarity parameter  $E_T(30)$  and the absorption maximum of (a) TPA-CN and (b) TPA-CNQ.

Solvent	$E_T(30)$ kcal mol <sup>-1</sup>	TPA-CN		TPA-CNQ		$E_g / eV^b$	
		Abs $\lambda_{max} / nm$	Em $\lambda_{max}$ /nm <sup>a</sup>	Abs $\lambda_{max} / nm$	Em $\lambda_{max} / nm^a$	TPA- CN	TPA- CNQ
Toluene	33.9	330, 412, 474	389 ( $\lambda_{ex} =$ 330)	394	449 ( $\lambda_{ex} = 394$ )	2.11	2.71
CHCl <sub>3</sub>	39.1	276, 330, 494	405 ( $\lambda_{ex} =$ 330)	308, 398, 650,	371, 431 ( $\lambda_{ex} = 308$ ) 463 ( $\lambda_{ex} = 398$ )	2.00	1.53
CH <sub>2</sub> Cl <sub>2</sub>	40.7	266, 328, 360, 490	410 ( $\lambda_{ex} =$ 328)	306, 398, 504, 622	360, 423 ( $\lambda_{ex} = 306$ ) 462 ( $\lambda_{ex} = 398$ )	2.04	1.65
DMF	43.2	292, 420, 476	439 ( $\lambda_{ex} =$ 292)	404, 750, 850	465 ( $\lambda_{ex} = 404$ )	2.06	1.40

Conditions: <sup>a)</sup> Emission peaks excited at wavelength absorption maximum; <sup>b)</sup> obtained from

UV-Vis spectra:  $E_g = 1240/\lambda_{max}$ .

**Table 1**  
SUMMARY OF UV-VIS AND  
FLUORESCENCE SPECTRA OF TPA-  
CN AND TPA-CNQ IN SOLVENTS

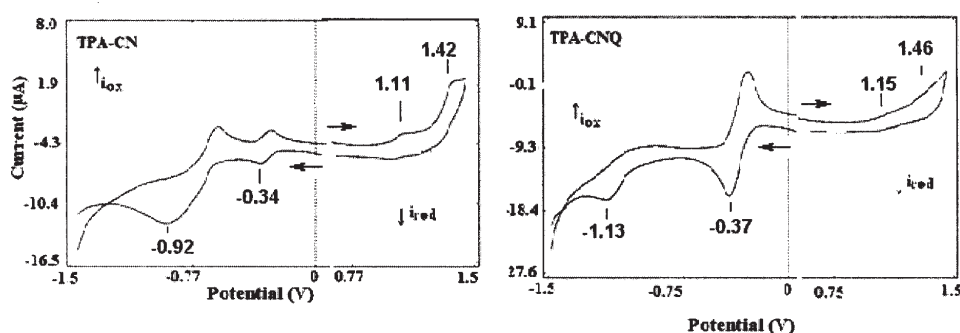


Fig.8. Cyclic voltammograms of compounds of oligomers in CH<sub>2</sub>Cl<sub>2</sub>. All solutions contained Bu<sub>4</sub>NF<sub>4</sub> (0.1M) as the supporting electrolyte, scan rate is 50 m V/s.

that these ICT molecules can absorb photons over a wide spectral range being interesting materials for solar cells.

As a conclusion, if the solvent polarity increased from nonpolar toluene to moderately polar (CHCl<sub>3</sub>, CH<sub>2</sub>Cl<sub>2</sub>) to highly polar DMF, the emission peaks are gradually red-shifted, exhibiting an obvious bathochromic effect.

#### Electrochemical studies

The electrochemical properties of oligomers with D-A structure are studied by cyclic voltammetry in dichloromethane, in a three-electrode electrochemical cell with Bu<sub>4</sub>NBF<sub>4</sub> (0.1M) as electrolyte and Ag/AgCl as reference electrode. Figure 8 shows typical CV curves of TPA-CN and TPA-CNQ, accounting for the reversible oxidation of TPA-group and reduction peaks ascribed to the =C(CN)<sub>2</sub> acceptor moieties. The two compounds exhibited in the both anodic and cathodic scan two reversible processes. In the negative potential region the compounds display reversible reduction processes, indicating formation of anions species. For TPA-CN, the oxidation peaks appearing at 1.11 V and 1.42 V being assigned to the formation of the TPA cation-radical and dications. Compound TPA-CNQ exhibited the same oxidation peaks at 1.15 V and 1.46 V, the second oxidation peak is not clear evidenced. The oxidation potential of the TPA-CNQ is more positive ( $E_{ox,1} = 1.15$  V) than that of TPA-CN ( $E_{ox,1} = 1.11$  V), which indicates that triphenylamine group is more electron-rich in TPA-CN.

The reversibility of oxidation and reduction processes of both D-A compounds shows that they could transport both electrons and holes being good ambipolar materials for organic field effect transistors.

#### Conclusions

Two new compounds of donor-acceptor type were synthesized starting from an activated alkyne and two strong electronoacceptor compounds (TCNE and TCNQ). These compounds displayed new absorption bands in the UV-Vis spectrum assigned to the intramolecular charge transfer transitions. Cyclic voltammetry has evidenced the ambipolar behaviour of the two compounds with reversible oxidation and reduction processes associated with presence of amine nitrogen and CN groups, respectively. The broad absorption responses over the solar spectrum and ambipolar character make these compounds interesting for electronics. Application of these compounds to organic solar cells and field effect transistors is also worthwhile for feature studies.

*Acknowledgements* : The authors thank to the Romanian National Authority for Scientific Research (UEFISCDI) for financial support (Grant PN-II-ID-PCE-2011-3-0274, Contract 148/2011).

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Manuscript received: 16.10.2012