

# New reactive hole-transporting hydrazone and its adducts with diol and dithiol

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New hole-transporting hydrazone with reactive vinyloxyethyl group and its glass forming adducts with 9-ethyl-3,6-dihydroxymethylcarbazole and 4,4'-thiobisbenzenethiol have been synthesized. The thermal stability of the synthesized compounds has been estimated and their optical, photophysical and photoelectrical properties have been studied. The ionisation potential of 5.14 eV has been established for 2-vinyloxyethoxy-4-diethylaminophenyl-1-carbaldehyde *N,N*-diphenylhydrazone by electron photoemission technique. Hole mobilities exceeding  $10^{-5}$  cm<sup>2</sup>/(V·s) at high electric fields have been observed by the time of flight technique in the 50 % solid solution of the hydrazone in bisphenol Z polycarbonate.

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## 1. Introduction

Among organic hole-transport materials used in electrophotographic photoreceptors aromatic amines and hydrazones prevail [1,2]. Aromatic hydrazones exhibit relatively high hole drift mobilities [3-7]. In addition, most of the reported hole-transporting hydrazones readily form glasses [6-7]. One such aromatic glass-forming hydrazone exhibited very high time of flight hole drift mobilities, which approached  $10^{-2}$  cm<sup>2</sup>/Vs at electric field of  $6.4 \times 10^5$  V/cm [5]. Hydrazones containing functional groups such as hydroxyl, epoxy are useful for the preparation of cross-linked and therefore solvent resistant electrophotographic layers [8]. Vinyl ethers are very reactive in cationic photopolymerizations. They can also be used for the preparation of polymers and copolymers by polyaddition [9] and for the synthesis of glass-forming molecular materials [10]. In electrophotographic photoreceptors charge-transporting compounds are usually used in the form of solid solutions in polymer hosts. The most widely used polymer hosts are polycarbonate and polyvinylbutyral. If the crystalline compounds are used as charge-transporting "guests" such systems are often morphologically unstable. Charge-transporting compounds tend to crystallize if their concentration in polymer hosts exceeds 50 %. This problem can be avoided either using amorphous charge-transporting "guests" or cross-linked systems.

Earlier we reported on the synthesis and properties of triphenylamine and carbazole based hydrazones having

vinyloxyethyl functional group at the hydrazone moieties [10,11]. The aim of this work was the synthesis and the study of the thermal, optical and photoelectrical properties of new vinyloxyethyl-substituted 2-hydroxy-4-diethylaminophenyl-1-carbaldehyde *N,N*-diphenylhydrazone and its adducts with diol and dithiol. In contrast to the earlier reported hydrazones [10,11] in this hydrazone vinyloxyethyl group is not linked to the hydrazone moiety. The availability of hydrazones containing vinyloxyethyl groups in different positions allow to compare their reactivities.

## 2. Experimental

### 2.1. Materials

9-Ethyl-9*H*-carbazole, 2-chloroethyl vinyl ether, *N*-phenylhydrazine, 4,4'-thiobisbenzenethiol, phosphorus oxychloride, *p*-toluenesulfonic anhydride, potassium carbonate, sodium borohydride, magnesium sulphate, sodium sulphate (all from Aldrich), potassium hydroxide, sodium chloride, (Lachema), triethylamine (Acros), were used as purchased without further purification. Organic solvents were purified and dried by the standard methods [12]. The synthesis of 9-ethyl-3,6-dihydroxymethylcarbazole was reported earlier [10].

*2-Hydroxy-4-diethylaminophenyl-1-carbaldehyde N,N-diphenylhydrazone* (**1**) 15.0 g (0.0776 mol) of 2-hydroxy-4-diethylaminophenyl-1-carbaldehyde were dissolved in 50 ml of methanol under mild heating. Then a

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solution of 25.69 g (0.1164 mol) of *N,N*-diphenylhydrazine hydrochloride in ca. 50 ml of methanol was added. The reaction mixture was stirred at ~ 30 °C for 2 h and cooled down. Then the reaction mixture was diluted with diethyl ether and washed with distilled water in a separatory funnel. The solution was dried using anhydrous sodium sulphate and filtered. The solvent was removed by rotary evaporation. The product was purified by column chromatography using an eluent mixture of hexane and chloroform in a volume ratio of 1:1 and crystallized from the eluent. The yield of C<sub>23</sub>H<sub>25</sub>N<sub>3</sub>O<sub>1</sub> (FW=359.38) was 74.73% (20.85 g of grey crystals); m.p. 100.5-101°C. IR (KBr windows) (in cm<sup>-1</sup>): ν(O-H) 3218, ν(C-H in Ar) 3057, ν(C-H) 2971; 2932; 2894, ν(C=C in Ar) 1633, 1596, 1495, ν(C-N) 1298, 1245, γ(Ar) 751, 700. MS (m/z): 360.4 (40%, M+1), 206.4, 120.1. <sup>1</sup>H NMR (100 MHz, CDCl<sub>3</sub>, δ, ppm): 1.15 (t, *J*=7.0 Hz, 6H, CH<sub>3</sub>); 3.33 (q, *J*=7.0 Hz, 4H, CH<sub>2</sub>); 6.73 (d, *J*=9.0 Hz, 1H, =CH); 7.05-7.45 (m, 13H, Ar), 11.25 (s, 1H, -OH).

*2-Vinyloxyethoxy-4-diethylaminophenyl-1-carbaldehyde N,N-diphenylhydrazone (2)* 7.0 g (0.0195 mol) of 2-hydroxy-4-diethylaminophenyl-1-carbaldehyde *N,N*-diphenylhydrazone (**1**) were dissolved in 20 ml of ethyl methyl ketone and 3.96 ml (0.0389 mol) of 2-chloroethyl vinyl ether were added. Then 2.18 g (0.0389 mol) of potassium hydroxide and 2.68 g (0.0195 mol) of potassium carbonate were added to the reaction mixture. The reaction mixture was refluxed for ca. 15 h and cooled down when no starting materials were left (TLC control). After cooling down the inorganic components were filtered off. The solvent was removed from the filtrate by rotary evaporation. The product was purified by column chromatography using an eluent mixture of hexane and acetone in a volume ratio of 7:1. The solvents were removed by rotary evaporation and the product was freeze dried. The yield of C<sub>27</sub>H<sub>31</sub>N<sub>3</sub>O<sub>2</sub> (FW=429.46) was 51.2% (4.28 g of yellowish brown resin). IR (KBr windows) (in cm<sup>-1</sup>): ν(C-H in Ar) 3060, 3023, ν(C-H) 2971; 2931; 2871, ν(C=C in Ar) 1698, ν(C-O-C) 1203, γ(Ar) 749, 700. MS (m/z): 430.4 (100%, M+1), 280.5. <sup>1</sup>H NMR (100 MHz, CDCl<sub>3</sub>, δ, ppm): 1.2 (t, *J*=7.0 Hz, 6H, CH<sub>3</sub>); 3.33 (q, *J*=7.0 Hz, 4H, CH<sub>2</sub>); 3.74-4.2 (m, 4H, -O-CH<sub>2</sub>), 6.07-6.09 (d, 2H, *J*=7.0 Hz, CH<sub>2</sub>=), 6.29 (q, *J*=7.0 Hz, 1H, O-CH=), 7.02-7.54 (m, 13H, Ar), 7.9 (d, *J*=8.0 Hz, 1H, -CH). Elemental analysis for C<sub>27</sub>H<sub>31</sub>N<sub>3</sub>O<sub>2</sub> (FW=429.46): calculated: C 75.49%; H 7.28%; N 9.78%; O 7.45%; experimentally obtained: C 75.19%; H 7.8%; N 9.51%.

*9-Ethyl-3,6-di-((2-((diphenylamino)iminomethyl)-5-(diethylamino)phenyloxyethoxy-1-methyl-methoxy)methyl)carbazole (3)* 1.0 g (0.0023 mol) of 2-Vinyloxyethoxy-4-diethylaminophenyl-1-carbaldehyde-*N,N*-diphenylhydrazone (**2**) and 0.27 g (0.0012 mol) of 9-ethyl-3,6-dihydroxymethylcarbazole were dissolved in 10 ml of dry tetrahydrofuran (THF) under nitrogen atmosphere. Then 1 ml of *p*-toluenesulphonic anhydride solution in dry THF (0.5×10<sup>-5</sup> mol) was added at the ambient temperature. The reaction mixture was stirred at the reflux temperature for

24 h. After that THF was evaporated. The crude product was purified by column chromatography using an eluent mixture of hexane and acetone in a volume ratio of 3:1. The solvent was removed with a rotary evaporator and the product was freeze dried. The yield of C<sub>70</sub>H<sub>79</sub>N<sub>7</sub>O<sub>6</sub> (FW=1114.17) was 15 % (0.17g of yellow resin). IR (KBr windows) (in cm<sup>-1</sup>): ν(C-H in Ar) 3060; 3023, ν(C-H) 2980, 2967, 2872, ν(C=C in Ar) 1611, 1598; 1516; 1495, ν(C-N) 1277, 1236, ν(C-O-C) 1213, γ(Ar) 805, 748, 700. <sup>1</sup>H NMR (100 MHz, CDCl<sub>3</sub>, δ, ppm): 1.01-1.31 (m, 15H, CH<sub>3</sub>), 1.37-1.72 (m, 10H, -CH<sub>2</sub>-N), 2.00-2.38 (m, 6H, CH<sub>3</sub>), 3.21-3.52 (m, 8H, -CH<sub>2</sub>-CH<sub>2</sub>-), 3.61-3.88 (d, 4H, *J*=6.0 Hz, O-CH<sub>2</sub>-Ar), 6.15 (s, 2H, -CH=), 6.29-6.50 (q, 2H, *J*=7.0 Hz, 2(-O-CH-O-), 7.02-8.19 (m, 32H, Ar).

*Di(4-(2-((diphenylamino)iminomethyl)-5-(diethylamino)phenyloxyethoxy-1-methyl-methylthio)phenyl)sulphide (4)* 1.0 g (0.0023 mol) of 2-vinyloxyethoxy-4-diethylaminophenyl-1-carbaldehyde-*N,N*-diphenylhydrazone (**2**) and 0.26g (0.0011 mol) of 4,4'-thiobisbenzenethiol were dissolved in 10 ml of dry THF under nitrogen atmosphere. Then 1 ml of a *p*-toluenesulphonic anhydride solution in dry THF (5×10<sup>-5</sup> mol) was added at the ambient temperature. The reaction mixture was stirred at the reflux temperature for 20 h until no changes in the reaction mixture were observed (TLC control). After that THF was evaporated. The crude product was purified by column chromatography using an eluent mixture of hexane and acetone in a volume ratio of 3:1. The solvent was removed with a rotary evaporator and the product was freeze dried. The yield of C<sub>66</sub>H<sub>72</sub>N<sub>6</sub>S<sub>3</sub>O<sub>4</sub> (FW=1109.28) was 22 % (0.25g of yellow resin). IR (KBr windows) (in cm<sup>-1</sup>): ν(C-H in Ar) 3061; 3024, ν(C-H) 2930, 2967, 2872, ν(C=C in Ar) 1611, 1516; 1498, ν(C-N) 1279, 1239, ν(C-O-C) 1213, γ(Ar) 808, 748, 701. <sup>1</sup>H NMR (100 MHz, CDCl<sub>3</sub>, δ, ppm): 0.82-1.09 (m, 12H, CH<sub>3</sub>), 1.20-1.31 (m, 8H, -CH<sub>2</sub>-N), 3.32-3.41 (m, 6H, CH<sub>3</sub>), 3.81-4.18 (t, 4H, *J*=5.0 Hz, -CH<sub>2</sub>-O-), 4.21-4.32 (t, 4H, *J*=5.0 Hz, -O-CH<sub>2</sub>-), 6.18 (s, 2H, -CH=), 6.34-6.52 (q, 2H, *J*=7.0 Hz, 2(-S-CH-O-), 6.90-8.28 (m, 34H, Ar).

## 2.2. Instrumentation

Infrared (IR) spectra were recorded using Bio-Rad Digilab FTS-40 and Perkin Elmer Spectrum GX spectrometers. The samples were prepared as KBr pellets.

Ultraviolet-visible (UV/VIS) spectra were recorded with Spectronic Unicam Genesys<sup>TM</sup> 8.

Fluorescence emission spectra were recorded with a Hitachi MPF-4 (Japan) luminescence spectrometer.

Proton nuclear magnetic resonance (<sup>1</sup>H NMR) spectra were obtained using a Bruker AC 250 (250 MHz), Varian Unity Inova (300 MHz) and JOEL FX 100 (100 MHz) apparatus. All the data are given as chemical shifts in δ (ppm), multiplicity, integration downfield from (CH<sub>3</sub>)<sub>4</sub>Si.

Mass (MS) spectra were obtained on Waters ZQ (Waters, Milford, USA).

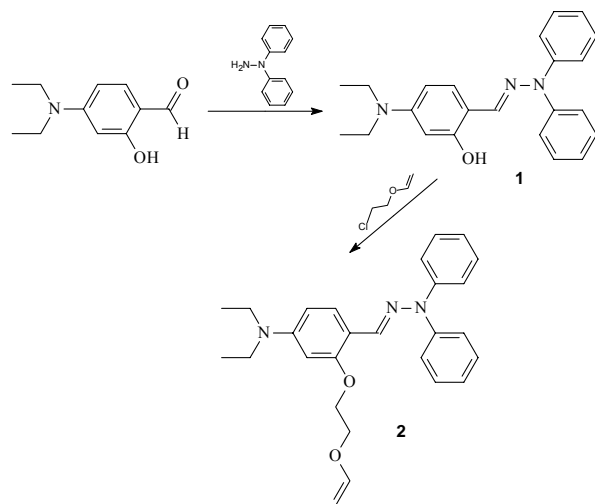
Thermogravimetric analysis (TGA) was performed on Netzsch STA 409 with a data acquisition system 414/1.

Differential scanning calorimetry (DSC) measurements were carried out using Perkin-Elmer DSC-7 calorimeter.

The ionisation potential ( $I_p$ ) was measured by the method of electron photoemission in air as reported earlier [13,14]. The samples for the measurements were prepared by casting the solutions of the compounds on Al plates pre-coated with an adhesive layer of methylmethacrylate and methacrylic acid copolymer. Hole-drift mobilities were measured by a time-of-flight method in a xerographic regime [15,16]. The samples for the measurements were prepared by casting the solutions in THF of the mixtures of the compounds with bisphenol Z polycarbonate (PC-Z) in mass proportion 1:1. The substrates were polyester films coated with thin Al layer. The thickness of the material casted on the substrate was estimated with a mechanical dial micrometer.

### 3. Results and discussion

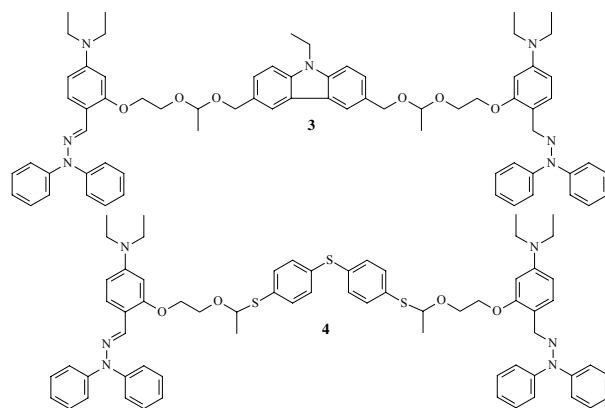
2-Vinyloxyethoxy-4-diethylaminophenyl-1-carbaldehyde *N,N*-diphenylhydrazone (**2**) was synthesized by the two-steps procedure as shown in Scheme 1. The first step was the condensation of 2-hydroxy-4-diethylaminophenyl-1-carbaldehyde with *N,N*-diphenylhydrazine hydrochloride and the second step was the alkylation of 2-hydroxy-4-diethylaminophenyl-1-carbaldehyde *N,N*-diphenylhydrazone (**1**) with 2-chloroethyl vinyl ether in the presence of KOH and  $K_2CO_3$ . All the products, were purified by crystallization followed by column chromatography. The synthesized materials were characterised by IR,  $^1H$  NMR spectroscopy, mass spectrometry and elemental analysis.



Scheme 1. Synthesis of 2-vinyloxyethoxy-4-diethylaminophenyl-1-carbaldehyde *N,N*-diphenylhydrazone (**2**).

Vinyloxyethyl-substituted hydrazone **2** was further used for the synthesis of hole-transporting amorphous

molecular materials. The structures of the synthesized materials are shown in Scheme 2. Compounds **3** and **4** were prepared by the reactions of compound **2** with 9-ethyl-3,6-dihydroxymethylcarbazole and respectively 4,4'-thiobisbenzenethiol in the presence of a catalytic amount of *p*-toluenesulphonic anhydride. The starting material 9-ethyl-3,6-dihydroxymethylcarbazole was synthesized by the reduction of 9-ethylcarbazole-3,6-dicarbaldehyde with sodium borohydride.



Scheme 2. Structures of 2-vinyloxyethoxy-4-diethylaminophenyl-1-carbaldehyde *N,N*-diphenylhydrazone-based molecular materials.

The signals in the  $^1H$  NMR spectra of all the newly synthesized hydrazone compounds can be exactly assigned to the characteristic hydrogen atoms of these compounds. The new characteristic signal at 6.73 ppm due to the proton of  $N=CH$  group appears in the spectrum of hydrazone **1**. The spectrum of compound **1** contains a well distinguished signal of OH proton at 11.25 ppm, which disappears completely in the spectrum of compound **2**. The signals of  $CH_2$  group protons of the ethyl vinyl ether moiety in the spectrum of the compounds **2** are observed at 3.74-4.2 ppm. The well distinguished signal of  $O-CH=$  proton is at 6.29 ppm in the spectrum of vinyloxyethyl substituted hydrazone. The signals at 7.02-7.90 ppm can be assigned to the aromatic and heterocyclic protons. The proton signals of double bond which are visible in the spectra of vinyloxyethyl-substituted hydrazone **2** at 6.07-6.09 ppm and at 6.29 ppm (proton signals of the group  $-O-CH=$ ) completely disappear in the spectra of compounds **3**, **4**.

Compounds **1-4** were also characterized by UV/VIS spectrometry. All these compounds absorb light in the region between 200 and 420 nm. The UV absorption spectra of dilute solutions of compounds **2-4** are given in Fig. 1. The spectra are rather similar. The lowest energy absorption band observed near 370 nm for compounds **3** and **4** exhibits hyperchromic effect with respect of that for

compound **2**. This observation can be explained by the higher content of hydrazone moieties in the molecules of hydrazones **3** and **4**. The UV spectrum of compound **3** shows K band of carbazole moiety (at ca.) near 300 nm, which is not characteristic for the spectra of compounds **2** and **4**.

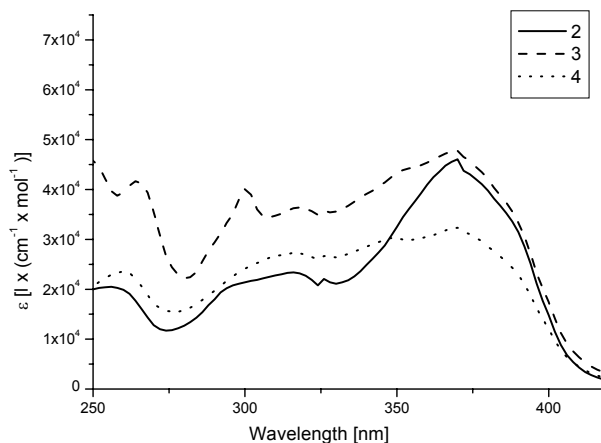


Fig. 1. UV absorption spectra of dilute THF solutions ( $10^{-5}$  M) of hydrazones 2-4.

The fluorescence emission spectra of the dilute solutions of compounds **2-4** are presented in Fig. 2. The fluorescence spectra do not depend on the excitation wavelength. All the compounds exhibit emission from the hydrazone moiety with the maxima at ca. 425 nm. It is interesting to note that no fluorescence from carbazole moiety is observed in the spectrum of **3**. This observation can be explained by the internal energy transfer from carbazole to hydrazone moiety.

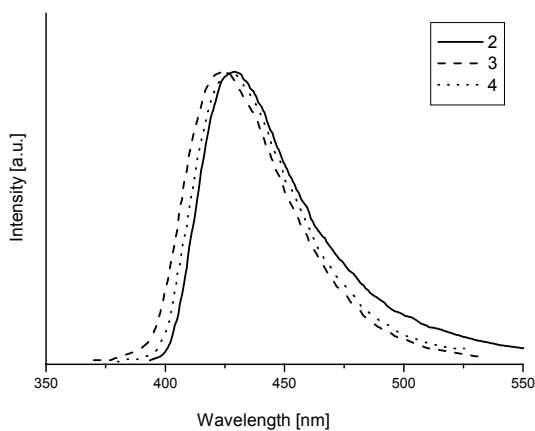


Fig. 2. Fluorescence spectra of dilute THF solutions ( $10^{-5}$  M) of hydrazones 2-4.

The thermal stability of hydrazones **2-4** was estimated by TGA. The thermal stability of aromatic hydrazones is

predetermined by the degradation of hydrazone moiety and usually does not exceed 300 °C [5]. The initial decomposition temperature ( $T_{ID}$ ) of compounds **2** and **3** is 240 °C and of hydrazone **4** is 265 °C. The thermal stability of these twin compounds depends on the origin of the linking bridge. Compound **3** containing flexible center di(thiophenyl)sulphide bridge exhibits a  $T_{ID}$  higher by 25 °C than the compound **4** containing 9-ethyl-3,6-oxymethylcarbazole bridge.

Compound **2** was isolated as a viscous resin. This material exhibited a glass transition at -3 °C. Compounds **3**, **4** were isolated as amorphous powders. Their films on substrates could be prepared by casting or spin coating techniques.

All the compounds described in this work are soluble in common organic solvents such as chloroform, acetone, THF. The ionisation potential of the film of hydrazone **2** was established by electron photoemission technique. The value of ionisation potential ( $I_p$ ) of hydrazone **2** is 5.14 eV. This value lower by ~ 0.3 eV than that earlier reported value for vinyl ethers of triphenylamine and carbazole based hydrazones [10,11]. The relatively low values of  $I_p$  make these compounds of interest for the application in electrophotographic photoreceptors. Holes would be injected easily into the layers of these materials from charge generation layers widely used in electrophotographic photoreceptors. The  $I_p$  values of charge generation materials used in electrophotographic photoreceptors such as titanil phthalocyaniones, perylene pigments and bisazo pigments are in the range 5.1-5.6 eV [13].

Fig. 3 shows the electric field dependencies of hole drift mobilities in the solid solutions of vinyloxyethyl-substituted hydrazone **2** and compounds **3**, **4** in bisphenol Z polycarbonate (PC-Z). The linear dependencies of the hole drift mobilities on the square root of the electrical field were observed for all the samples.

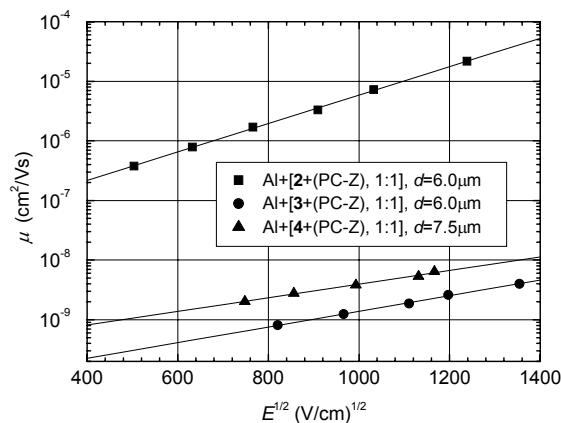


Fig. 3. Electric field dependencies of holes drift mobilities in the amorphous films of the 50 % solid solutions of hydrazones 2-4 in PC.

The highest holes drift mobilities were observed for the solid solutions of **2** in PC-Z. The hole drift mobilities of this system exceed  $10^{-5}$  cm<sup>2</sup>/(V·s) at high electric fields.

These are relatively high mobilities as for molecularly doped polymers. In the films of molecular glasses charge mobilities are usually by 1-2 orders of magnitude higher than in molecularly doped polymers containing 50 % of the active material [2]. The solid solutions of compounds **3** and **4** in PC-Z showed much lower mobilities. At high electrical fields they hardly reach  $10^{-8}$  cm<sup>2</sup>/(V·s). Lower charge mobilities in the systems containing **3** and **4** relative to the system containing **2** can be explained by the lower concentration of electrophores in these systems. Poor charge transport properties of compounds **3** and **4** can apparently be explained by unfavourable packing of big non-planar molecules for the electron transfer from the neutral molecules to the radical cations.

#### 4. Conclusions

New hole-transporting hydrazone with reactive vinyloxyethyl group has been synthesized by two steps procedure. The reactions of newly synthesized 2-vinyloxyethyloxy-4-diethylaminophenyl-1-carbaldehyde N,N-diphenylhydrazone with 9-ethyl-3,6-dihydroxy-methylcarbazole and 4,4'-thiobisbenzenethiol yielded new-hole transporting amorphous molecular materials. The thermal stability of the synthesized compounds has been estimated and their optical, photophysical and photoelectrical properties have been studied. The ionisation potential of 5.14 eV has been established for 2-vinyloxyethyloxy-4-diethylaminophenyl-1-carbaldehyde N,N-diphenylhydrazone by electron photoemission technique. Hole mobilities exceeding  $10^{-5}$  cm<sup>2</sup>/(V·s) at high electric fields have been observed by the time of flight technique in the 50 % solid solution of the hydrazone in bisphenol Z polycarbonate.

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