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Di(9-alkylcarbazol-3-yl)arylamines as Electroactive Amorphous Materials for Optoelectronics

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Summary. Star-shaped molecules of di(9-alkylcarbazol-3-yl)arylamines were synthesized and found to constitute new glass-forming materials with glass transition temperatures ranging from 40 to 147°C. The electron photoemission spectra of the molecular glasses were recorded and ionisation potentials of 4.9–5.0 eV were established. Room temperature time of flight hole drift mobilities of the di(9-alkylcarbazol-3-yl)phenylamines molecularly dispersed in polycarbonate-Z approached $5 \cdot 10^{-6}\,\mathrm{cm^2/Vs}$ at high electric fields. Some of the compounds were converted to cross-linkable derivatives, which are potential components for insoluble charge transport layers.

Keywords. Aromatic amine; Molecular glasses; Ionisation potential; Charge transport.

Introduction

Film-forming electroactive organic materials are known for various applications such as electrophotographic photoreceptors [1], organic light emitting diodes [2, 3], photovoltaic cells [4], photorefractive materials [5], and organic transistors [6]. Amorphous organic electroactive materials are divided into two categories. One is low-molar-mass molecules; the devices are generally fabricated by vacuum-deposition. The other is polymers; the devices are fabricated by the casting or spin-coating methods. Among the low-molar-mass derivatives, triphenylamine, triphenyldiamine (*TPD*), and their derivatives are the most extensively used as hole transport materials. They are easily oxidized to form stable radical cations [7, 8]. However, the low-molar-mass triphenylamine and *TPD* derivatives have some shortcomings, such as low morphological stability of their amorphous films as well as rather high ionization potentials [9, 10]. We have reported previously on carbazol-9-yl substituted *TPD* derivatives which formed amorphous films with glass transition

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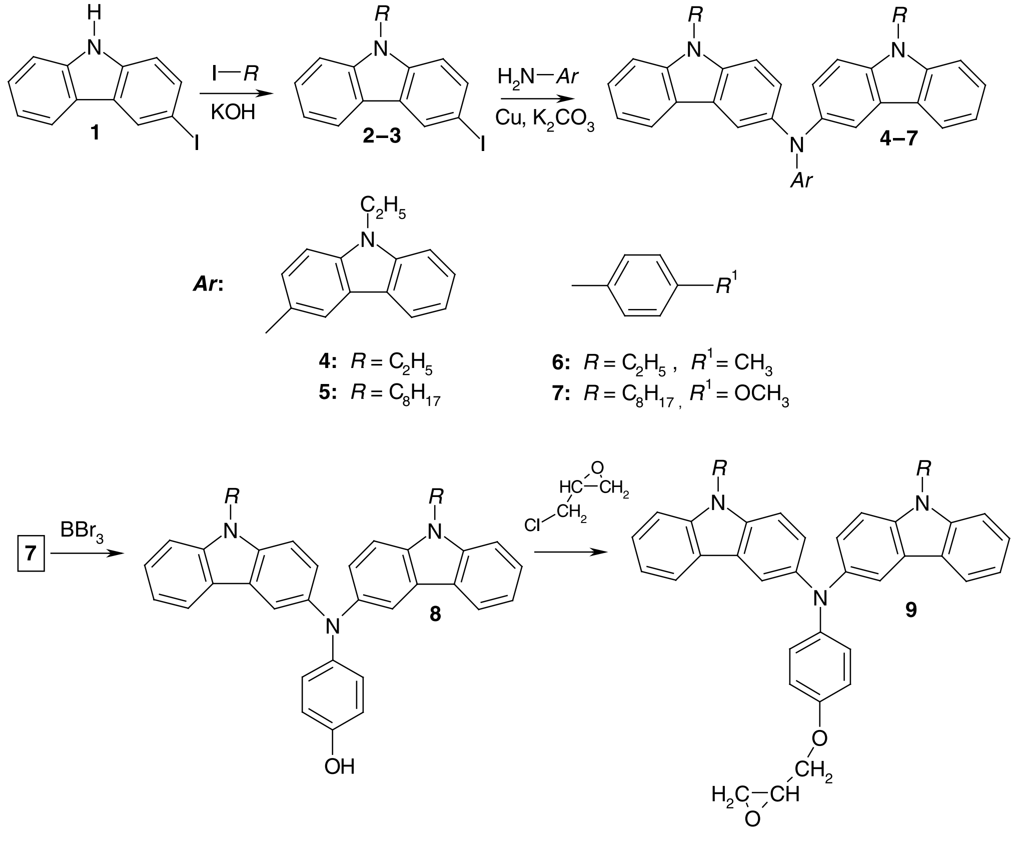
temperatures (T_g) higher than 100°C [11]. Electron photoemission spectroscopy of thin films of the compounds showed ionisation potentials of *ca.* 5.4 eV.

In this work, we have designed and synthesised new triarylamines having the bulky carbazolyl species, which enhances the amorphous nature of low-molar-mass compounds. These branched structures are expected to form amorphous films with good morphological stability due to the geometry of these molecules, which does not favour close packing. We also report on the synthesis and properties of cross-linkable triarylamine derivatives, which could be applied in manifold ways for optoelectronic devices [12–14]. Photo-cross-linking is a useful method of protecting thin organic systems against solvents [15, 16].

Results and Discussion

Di(9-alkylcarbazol-3-yl)arylamines **4**–**7** were generally prepared as described in Scheme 1 *via* the modified *Ullmann* coupling reaction [17, 18] of an excess of 3-iodo-9-alkylcarbazole **2** or **3** with 3-amino-9-ethylcarbazole, 4-toluidine, or 4-anisidine. The iodo-derivatives **2** and **3** were prepared by alkylation of 3-iodo-9*H*-carbazole (**1**) which was synthesized by iodination of 9*H*-carbazole.

N-(4-Methoxyphenyl)-*N*,*N*-di[9-(2-ethylhexyl)carbazol-3-yl]amine (**7**) was used for the preparation of the reactive di(9-alkylcarbazol-3-yl)phenylamines **8** and **9** by



Scheme 1

Table 1. Thermal characteristics of **4–8**

Compound	T_g/°C	T_m/°C	T_{ID}/°C
4	147	323	432
5	66	–[a]	444
6	40	–[a]	367
7	39	–[a]	404
8	48	111	369

[a] Obtained as an amorphous substance

two-step reaction as shown in Scheme 1. The first step involved cleavage of the methyl hydroxy-protecting group using BBr_3 [19] to get **8**. The second step was condensation of the hydroxyl substituted aromatic amine **8** with epichlorohydrin in the presence of benzyltrimethylammonium cations.

All the new compounds were identified by mass spectrometry, IR, and electronic absorption, as well as ^{1}H NMR spectroscopy. The data were found to be in good agreement with the proposed structures. The di(9-alkylcarbazol-3-yl)arylamines **4**–**9** were soluble in common organic solvents, such as tetrachloroethane, $CHCl_3$, *THF*, and acetone at room temperature. Transparent and stable thin amorphous films of these materials could be prepared by casting or spin coating from solutions.

The behaviour of **4**–**8** under heating was studied by DSC and TGA under N_2 atmosphere. The values of T_g, melting points (T_m), and the temperatures at which initial loss of mass was observed (T_{ID}) are summarised in Table 1. All the derivatives demonstrate high thermal stability. The mass loss occurs at temperatures higher than 367°C as confirmed by TGA with a heating rate of 10°C/min.

It is evident that the thermal stability of **4**–**8** decreases with the introduction of a 4-substituted phenyl group (*cf.* T_{ID} of **4** and **5** with that of **6**–**8**). T_{ID} only slightly depends on the substituent at the *p*-position of the phenyl group (*cf.* T_{ID} of **7** with that of **6** and **8**).

Derivatives **4** and **8** were obtained as polycrystals by crystallisation from solutions, however, they readily formed glasses when the melt samples were cooled on standing in air or with liquid N_2. The DSC thermograms of **4** are shown in Fig. 1a. When the crystalline sample was heated, the endothermic peak due to melting was observed at 323°C. When the melted sample was cooled down and heated again, the glass-transition phenomenon was observed at 147°C and on further heating no peaks due to crystallisation and melting appeared. The crystalline sample of **8** demonstrated a similar behaviour. It melts on first heating at 111°C and forms a glass ($T_g = 48$°C) upon cooling.

Compounds **5**–**7** were obtained as amorphous materials as confirmed by DSC. As an example, the DSC thermograms of **5** are shown in Fig. 1b. When a sample of **5** was heated T_g was observed at 66°C, and no peaks due to crystallisation and melting appeared. Cooling down and repeated heating revealed the glass transition again only.

UV absorption and emission spectra of dilute solutions of **4**–**9** were recorded. Selected spectra are presented in Fig. 2. For comparison the corresponding spectra of 9-ethylcarbazole are given. The carbazole trimers **4** and **5** as well as the series of

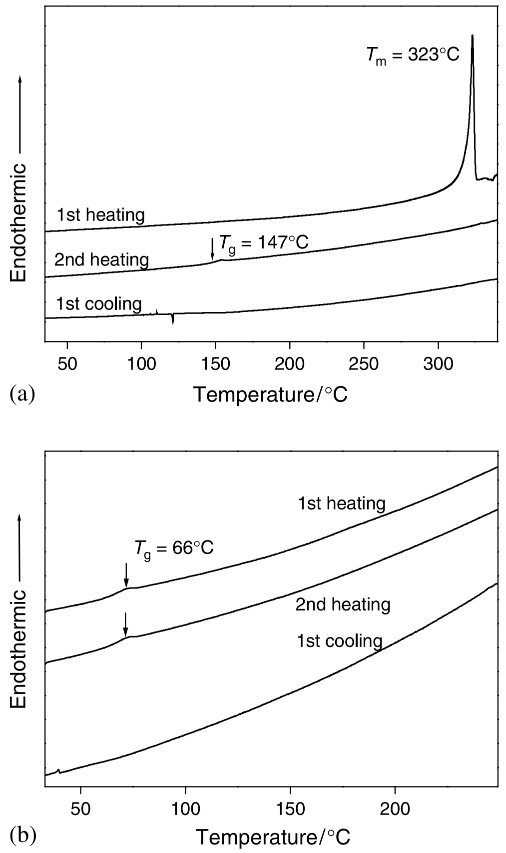


Fig. 1. DSC curves of **4** (a) and **5** (b); heating rate 10°C/min

di(9-alkylcarbazol-3-yl)phenylamines **6**–**9** exhibit very similar UV and fluorescence spectra, therefore only the spectra of **5** and **6** are shown. The lowest energy absorption maxima of these compounds exhibit bathochromic shifts with respect to the corresponding absorption maxima of 9-ethylcarbazole. This circumstance influences fluorescence spectra, which are considerably red-shifted with respect of the fluorescence spectrum of 9-ethylcarbazole. The emission maxima of **4**–**9** appear in the region from 448 to 466 nm.

The red shifts of UV absorption and emission, which are interpreted as extended conjugation, stabilizing the oxidized state of the compounds, are expected to influence the values of ionization potentials (I_p).

The electron photoemission spectra of **5** and **6** are presented in Fig. 3. For comparison the spectrum of poly(vinylcarbazole) (*PVK*) containing unsubstituted carbazole rings as chromophores is shown. The values of I_p are 4.9 for **4** and **5**, *ca.* 5.0 for **6**–**9**, and 5.77 eV for *PVK*. Thus, the I_ps of the newly synthesized compounds are much lower than those of *PVK* and other polymers having electronically isolated carbazole electrophores [20].

Holes would be easily injected into the layers of compounds **4**–**9** from a conductive anode or charge generation layer with a work function or I_p close to 5.0 eV. The I_p values for charge generation materials, including those widely used in electrophotographic photoreceptors, *e.g.* titanyl phthalocyanines [21, 22], perylene

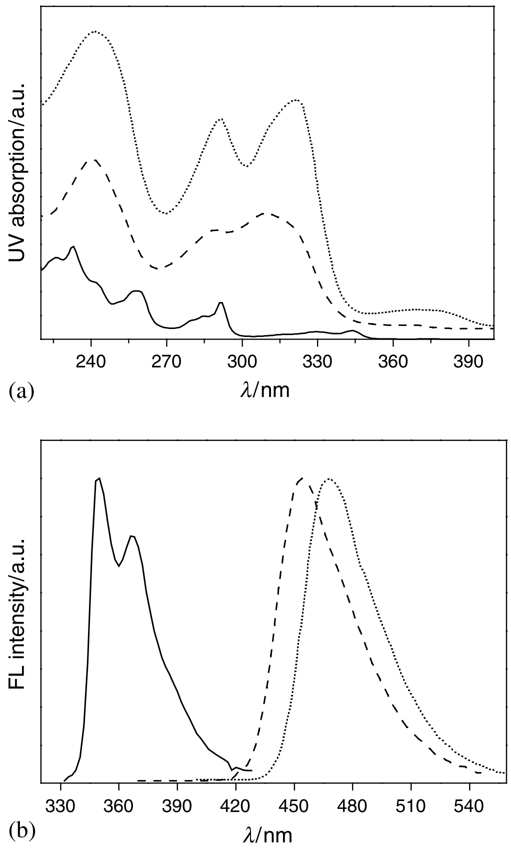


Fig. 2. UV absorption (a) and PL emission (b) spectra of dilute *THF* solutions of **5** (dotted line), **6** (dashed line), and 9-ethylcarbazole (solid line)

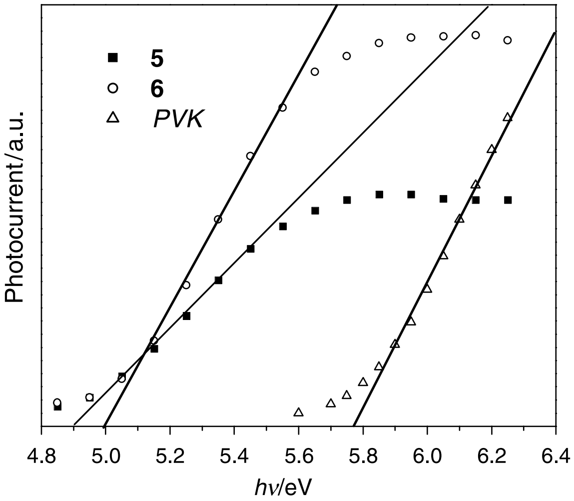


Fig. 3. The electron photoemission spectra of **5**, **6**, and *PVK*

pigments [23], and bisazo pigments [24] are in the range of 5.1–5.6 eV. The I_p values of **4**–**9** are very close to that of indium-tin oxide (4.8 eV), which is widely used as an anode in electroluminescent devices [25]. The injection barrier of holes from the electrode into the layers of the materials synthesized would be only 0.1–0.2 eV.

Compounds **6** and **7** having the best solubility in PC-Z solutions were used for charge transport studies. Xerographic time of flight (XTOF) measurements were used to characterize the magnitudes of the hole drift mobility (μ_h) for the compounds molecularly dispersed in PC-Z. Transit time for the samples with the transporting materials was determined by a kink on the dU/dt transient in double logarithmic scale (Fig. 4).

The room temperature μ_h showed a linear dependence on the square root of the electric field (Fig. 5). Such dependence is observed for the majority of non-crystalline organic systems and is attributed to the effects of disorder on the charge transport [26]. Solid solutions of **6** or **7** in PC-Z (50%) demonstrated hole drift mobility values reaching $2 \cdot 10^{-6}\,\mathrm{cm^2/Vs}$ at $8.1 \cdot 10^5\,\mathrm{V/cm}$.

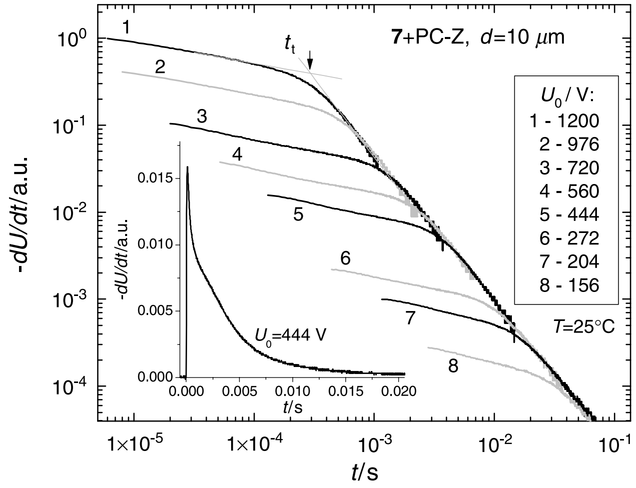


Fig. 4. XTOF transients ot **7** doped in PC-Z (50%); insert shows one of the transient curves in linear plot

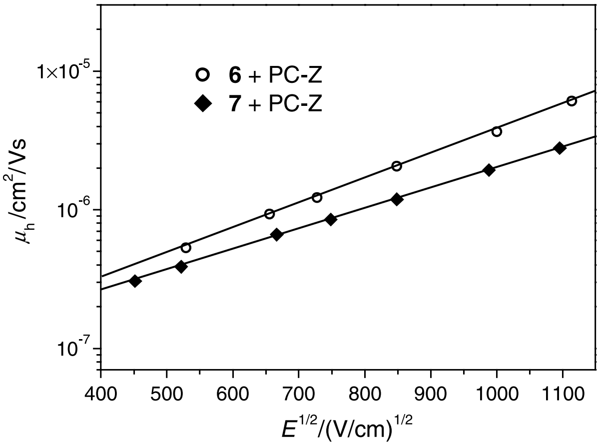


Fig. 5. The electric field dependencies of the hole drift mobility in charge transport layers of **6** and **7** doped in PC-Z

In conclusion, we have synthesized a series of di(9-alkylcarbazol-3-yl)arylamine based compounds which exhibit high thermal stability and form amorphous films with glass transition temperatures in the range of 40–147°C. Some of the compounds were converted into reactive derivatives, which are potential components for the preparation of insoluble electroactive layers. The values of ionisation potentials (4.9–5.0 eV) and the results of hole drift mobility studies have shown that these compounds are potential materials for electrophotographic photoreceptors and multilayer electroluminescent devices.

Materials and Methods

^{1}H NMR spectra were recorded using the Varian Unity Inova and JOEL JNM-FX 100 apparatuses. Mass spectra were obtained on a Waters ZQ 2000 spectrometer. FTIR spectra were recorded using Perkin Elmer FT-IR System. UV spectra were measured with a Spectronic Genesys^{TM} 8 spectrometer. Fluorescence spectra were recorded with a MPF-4 spectrometer. DSC measurements were carried out using a Perkin Elmer Pyris Diamond calorimeter. TGA was performed on a Netzsch STA 409. The TGA and DSC curves were recorded in a N_2 atmosphere at a heating rate of 10°C/min.

The ionisation potentials of the layers of **4**–**9** were measured by the electron photoemission method in air described earlier [27, 28].

The hole drift mobility was measured by the xerographic time of flight technique [29, 30]. The samples for the charge carrier mobility measurements were prepared by casting solutions of the mixtures of **6** and **7** with bisphenol Z polycarbonate (PC-Z) in weight proportion 1:1 on polyester films with Al layer. The thickness of the charge transport layers varied in the range of 4–10 μm. Positive corona charging created electric field in the sample. Charge carriers were generated at the layer surface by illumination with pulses of UV flash (pulse duration was 1 μs, strong absorption). The layer surface potential decrease as a result of pulse illumination was up to 2–5% of the initial potential before illumination. The capacitance probe measured the rate of the surface potential decrease dU/dt. The transit time t_t was determined by the kink on the curve of the dU/dt transient in double logarithmic scale. The drift mobility was calculated by the formula $\mu_h = d^2/U_0 t_t$, where d is the layer thickness and U_0 is the surface potential at the moment of illumination.

9*H*-Carbazole, 3-amino-9-ethylcarbazole, 4-anisidine, 4-toluidine, alkyliodides, and epichlorohydrin were purchased from Aldrich and used as received. 3-Iodo-9*H*-carbazole (**1**) was obtained by a modified procedure of *Tucker* [31]. 3-Iodo-9-alkylcarbazoles **2** and **3** were prepared by alkylation of **1** in the presence of a phase transfer catalyst [32].

N,N,N-Tris(9-ethylcarbazol-3-yl)amine (**4**, $C_{43}H_{39}N_4$)

A mixture of 2 g (10.2 mmol) 3-amino-9-ethylcarbazole, 15.6 g (50.8 mmol) 3-iodo-9-ethylcarbazole (**2**), powdered K_2CO_3 (11.2 g, 81.5 mmol), Cu powder (2.64 g, 40.9 mmol), and 18-crown-6 (0.53 g, 2 mmol) were refluxed in 20 cm^3 *o*-dichlorobenzene under N_2 for 24 h. Cu and inorganic salts were filtered and the solvent was removed by distillation. The crude product was purified by silica gel column chromatography using ethyl acetate/*n*-hexane (*v*/*v* 1/4) as eluent. The product was crystallized from the eluent to yield 1.2 g of yellow crystals (mp 323°C). IR (KBr): $\bar{\nu} = 3050$ (CH_{ar}), 2973, 2981, 2890 ($CH_{aliphatic}$), 1627, 1601, 1482, 1470, 1460 ($C{=}C_{ar}$) cm^{-1}; MS (70 eV): $m/z = 597$ (M^+), 403; ^1H NMR (300 MHz, $CDCl_3$): $\delta = 1.49$ (t, $3CH_3$, $J = 4.0$ Hz), 4.44 (q, $3CH_2$–N, $J = 8.0$ Hz), 7.25–8.43 (m, $21CH_{ar}$) ppm.

N-(9-Ethylcarbazol-3-yl)-N,N-di[9-(2-ethylhexyl)carbazol-3-yl]amine (**5**, $C_{54}H_{60}N_4$)

Prepared from 1 g (2.6 mmol) 3-amino-9-ethylcarbazole and 9.9 g (13.2 mmol) 3-iodo-9-(2-ethylhexyl)carbazole (**3**) using powdered K_2CO_3 (5.4 g, 21.1 mmol), Cu powder (1.23 g, 19.4 mmol), and 18-crown-6 (0.26 g, 1 mmol) in the same way as for **4**. The compound was purified by column chromatography with silica gel using ethyl acetate/*n*-hexane (*v*/*v* 1/5) as eluent. Yield 1 g amorphous yellow powder. IR (KBr): $\bar{\nu} = 3049$ (CH_{ar}), 2957, 2928, 2871, 2858 ($CH_{aliphatic}$), 1627, 1601, 1464, 1458 ($C{=}C_{ar}$) cm^{-1}; MS (70 eV): $m/z = 765$ (M^+), 113, 167; ^1H NMR (300 MHz, *DMSO*-d_6): $\delta = 0.78$ (t, $2CH_3$, $J = 6.6$ Hz), 0.86 (t, $2CH_3$, $J = 7.0$ Hz), 1.17 (t, CH_3, $J = 4.8$ Hz), 1.20–1.33 (m, $8CH_2$), 1.92–2.00 (m, 2CH), 2.51–2.53 (m, $2CH_2$), 4.19 (d, $2CH_2$–N, $J = 7.3$ Hz), 4.38 (q, CH_2–N, $J = 7.0$ Hz), 6.97–7.84 (m, $21CH_{ar}$) ppm.

N-(4-Methylphenyl)-N,N-di[9-(2-ethylhexyl)carbazol-3-yl]amine (**6**, $C_{47}H_{55}N_3$)

A mixture of 1 g (9.45 mmol) *p*-toluidine, 11.5 g (28.34 mmol) 3-iodo-9-(2-ethylhexyl)carbazole (**3**), powdered K_2CO_3 (10.4 g, 75.6 mmol), Cu powder (2.4 g, 37.8 mmol), and 18-crown-6 (0.5 g, 1.9 mmol) were refluxed in 10 cm^3 *o*-dichlorobenzene under N_2 for 20 h. Cu and inorganic salts were filtered off and the solvent was removed by distillation. The residual product was purified by silica gel column chromatography using $CHCl_3$/*n*-hexane (*v*/*v* 1/5) as eluent. After separation the product was dissolved in *THF* and precipitated into methanol to provide 3 g of an amorphous yellow powder. IR (KBr): $\bar{\nu} = 3051$, 3023 (CH_{ar}), 2957, 2928, 2871, 2857 ($CH_{aliphatic}$), 1628, 1611, 1486, 1465 ($C{=}C_{ar}$) cm^{-1}; MS (70 eV): $m/z = 662$ (M^+); ^1H NMR (300 MHz, $CDCl_3$): $\delta = 0.93$ (t, $2CH_3$, $J = 6.8$ Hz), 0.99 (t, $2CH_3$, $J = 7.3$ Hz), 1.30–1.50 (m, $10CH_2$), 2.09–2.16 (m, 2CH), 2.22 (s, CH_3), 7.18–7.99 (m, $18CH_{ar}$) ppm.

N-(4-Methoxyphenyl)-N,N-di[9-(2-ethylhexyl)carbazol-3-yl]amine (**7**, $C_{47}H_{55}N_3O$)

Prepared from 2.6 g (20.78 mmol) *p*-anisidine and 25.25 g (62.33 mmol) **3** using powdered K_2CO_3 (22.9 g, 166.2 mmol), Cu powder (5.2 g, 83.1 mmol), and 18-crown-6 (1.1 g, 4.2 mmol) in the same way as for **6**. The compound was purified by column chromatography with silica gel using ethyl acetate/*n*-hexane (*v*/*v* 1/5) as eluent. After separation the product was dissolved in *THF* and precipitated into methanol to provide 6 g of an amorphous yellow powder. IR (KBr): $\bar{\nu} = 3049$ (CH_{ar}), 2956, 2928, 2870, 2857 ($CH_{aliphatic}$), 1627, 1601, 1482, 1465 ($C{=}C_{ar}$), 1239 (C–O–C) cm^{-1}; MS (70 eV): $m/z = 678$ (M^+), 113; ^1H NMR (300 MHz, *DMSO*-d$_6$): $\delta = 0.77$ (t, 2CH$_3$, $J = 7.0$ Hz), 0.85 (t, 2CH$_3$, $J = 7.0$ Hz), 1.16–1.34 (m, 8CH$_2$), 1.92–2.0 (m, 2CH), 3.69 (s, O–CH$_3$), 4.17–4.19 (d, 2CH$_2$–N, $J = 5.9$ Hz), 6.78–7.90 (m, 18CH$_{ar}$) ppm.

N-(4-Hydroxyphenyl)-N,N-di[9-(2-ethylhexyl)carbazol-3-yl]amine (**8**, $C_{46}H_{53}N_3O$)

N-(4-Methoxyphenyl)-*N*,*N*-di[9-(2-ethylhexyl)carbazol-3-yl]amine (**7**) (3 g, 4.4 mmol) was dissolved in 27 cm^3 dry CH_2Cl_2 at room temperature and cooled to −80°C under N_2. Boron tribromide (22 cm^3 of 1 *M* CH_2Cl_2 solution, 22.1 mmol) was added slowly to the mixture and it was stirred at −80°C for 30 min and at room temperature for 12 h. The reaction mixture was poured into H_2O and the product was extracted with $CHCl_3$. The organic phase was dried (Na_2SO_4), filtered, and concentrated under vacuum. The crude product was purified by silica gel column chromatography with silica gel using ethyl acetate/*n*-hexane (*v*/*v* 1/10) as eluent. The product **8** was crystallized from the eluent to yield 1.16 g of green crystals (mp 111°C). IR (KBr): $\bar{\nu} = 3502$ (OH), 3049 (CH$_{ar}$), 2957, 2927, 2870, 2858 (CH$_{aliphatic}$), 1627, 1600, 1483, 1465 (C=C$_{ar}$) cm^{-1}; MS (70 eV): $m/z = 664$ (M$^+$), 391, 149; ^{1}H NMR (300 MHz, *DMSO*-d$_6$): $\delta = 0.78$ (t, 2CH$_3$, $J = 7.3$ Hz), 0.86 (t, 2CH$_3$, $J = 7.3$ Hz), 1.17–1.35 (m, 8CH$_2$), 1.94–2.02 (m, 2CH), 3.36 (d, 2CH$_2$, $J = 9$ Hz), 4.20 (s, OH), 6.69–9.14 (m, 18CH$_{ar}$) ppm.

N-[4-(2,3-Epoxypropyl)oxyphenyl]-N,N-di[9-(2-ethylhexyl)carbazol-3-yl]amine (**9**, $C_{49}H_{57}N_3O_2$)

N-(4-Hydroxyphenyl)-*N*,*N*-di[9-(2-ethylhexyl)carbazol-3-yl]amine (**8**) (0.95 g, 1.43 mmol), epichlorohydrin (4.3 cm^3), and 0.2 g benzyltrimethylammonium chloride were refluxed for 1 h. The excess of epichlorohydrin was removed by distillation and the crude product was dissolved in $CHCl_3$ and washed with H_2O. Purification of **9** was achieved by silica gel column chromatography using acetone/*n*-hexane (*v*/*v* 1/15) as eluent. Yield 0.3 g of brown oil. IR (KBr): $\bar{\nu} = 3053$ (CH$_{ar}$), 2959, 2929, 2870, 2872 (CH$_{aliphatic}$), 1482, 1465 (C=C$_{ar}$), 1217 (C–O–C) cm^{-1}; MS (70 eV): $m/z = 720$ (M$^+$), 149; ^{1}H NMR (300 MHz, $CDCl_3$): $\delta = 0.91$ (t, 2CH$_3$, $J = 7$ Hz), 0.97 (t, 2CH$_3$, $J = 7.3$ Hz), 1.29–1.45 (m, 10CH$_2$), 2.05–2.15 (m, 2CH), 2.61 (d, CH$_2$, $J = 8.0$ Hz), 2.93–2.96 (m, 2CH), 3.41 (d, CH$_2$, $J = 5.9$ Hz), 6.75–8.24 (m, 18CH$_{ar}$) ppm.

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