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Synthesis and Properties of Glass-Forming Hydrazones II [1]. Hydrazones Containing Bicarbazolyl Units

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Summary. A series of new glass-forming hydrazones containing bicarbazolyl units were synthesized starting from 9-(2-(9*H*-carbazol-9-yl)-cyclobutyl)-9*H*-carbazole and 9-(6-(9*H*-carbazol-9-yl)-hexyl)-9*H*-carbazole, and their thermal properties were studied. The correlation between their inclination to form glasses and their chemical structures are discussed. The results of a preliminary investigation of the photoelectric properties of amorphous films of the title compounds are briefly reported. The highest hole drift mobility was observed for 9-(2-(9*H*-carbazol-9-yl)-cyclobutyl)-9*H*-carbazole-3-carbaldehyde N,N-diphenylhydrazone; at high electric fields, it approaches $1 \cdot 10^{-3}\,\mathrm{cm}^2/\mathrm{Vs}$.

Keywords. Heterocycles; Molecular glasses; Ionization potential; Charge transport.

Introduction

Hydrazones are known as very effective charge-transporting compounds. Together with polymer binders they are used in electrophotographic photoreceptors [2–6]. Most of the hydrazones used in electrophotography are crystalline compounds of low molar mass. The amount of such a hydrazone in a polymer binder usually cannot exceed 50% because of its tendency to crystallize. The use of amorphous hydrazone compounds apparently allows to increase their concentration in a polymer binder and thus to increase the charge carrier mobility in charge transport layers of electrophotographic materials. Among molecular glasses which have recently been widely studied in the context of application in electroluminescent devices and photorefractive materials, only few hydrazones are reported [7–10]. In a previous paper we have reported on the synthesis and glass-forming properties of carbazole-based dihydrazones [1]. Such dihydrazones can be transformed into a glassy state; however, the morphological stability of most of the glasses has been found to be too low. We have shown that introduction of the rigid phenyl group into the position 9 of the carbazole ring markedly increases the stability of the glasses. Here we report on the synthesis and properties of 9-(6-(9*H*-carbazol-9-yl)-hexyl)-9*H*-carbazole (*CHC*)- and 9-(2-(9*H*-carbazol-9-yl)-cyclobutyl)-9*H*-carbazole (*CCBC*) based hydrazones. Increase of the molar mass usually leads to increasing stability

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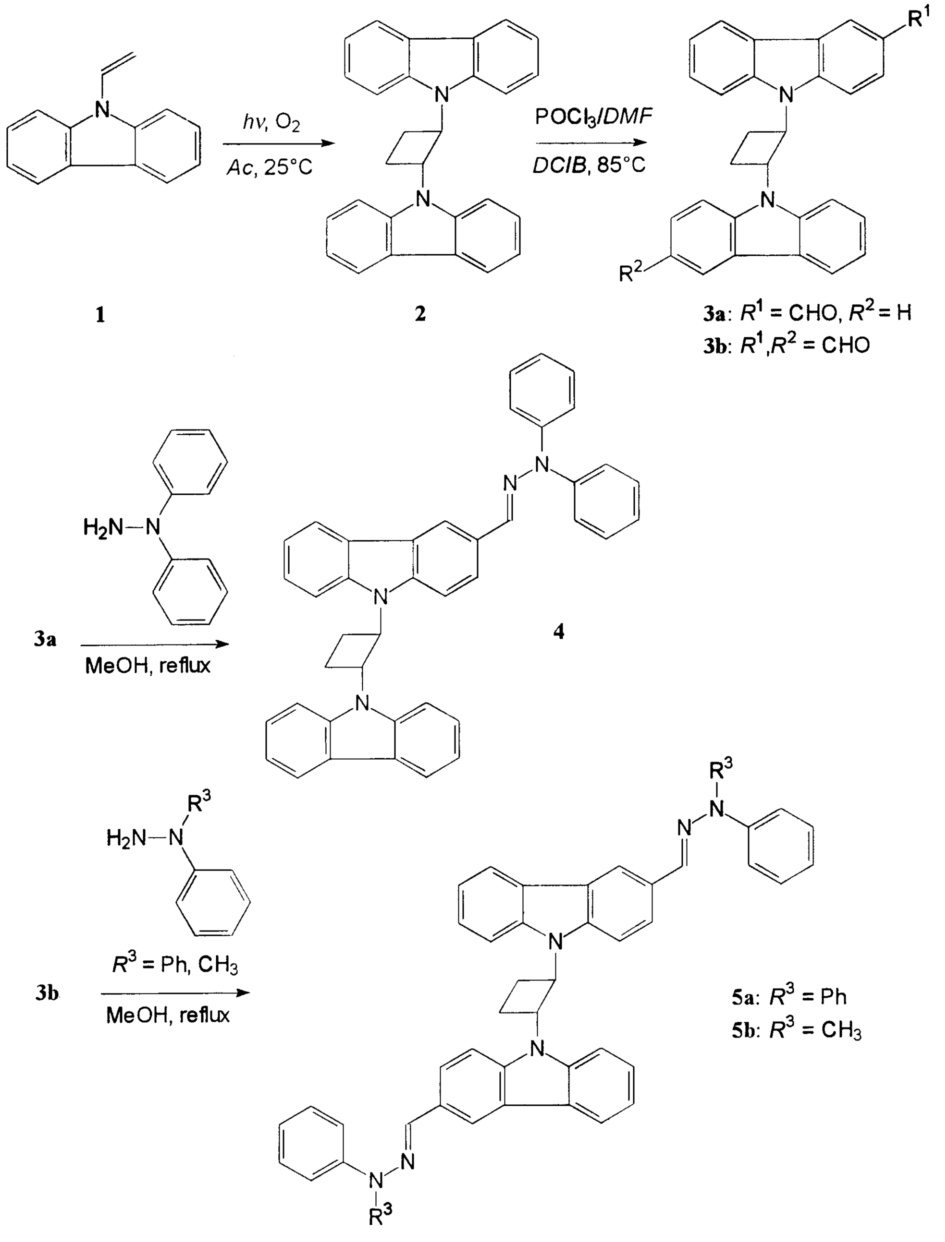
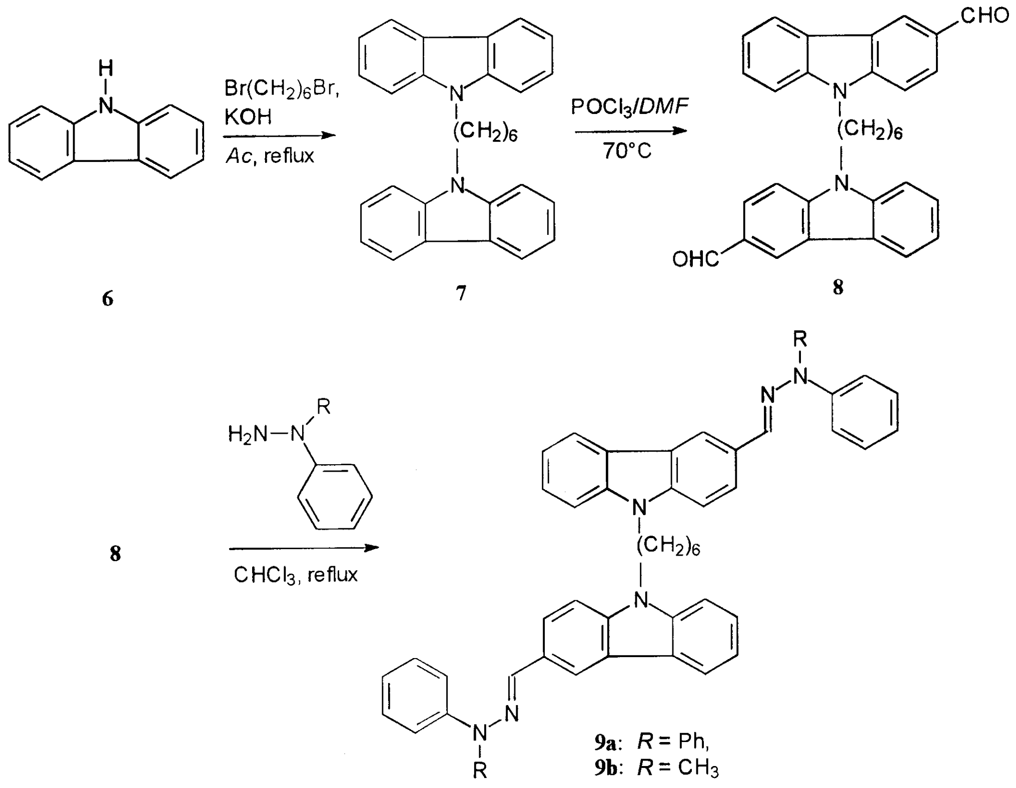
of molecular glasses [11]. In addition, *CCBC* is known to be a nonplanar moiety [12, 13]. This circumstance should also enhance the inclination of *CCBC* based molecules to form glasses.

Results and Discussion

CCBC based dihydrazones were prepared by the three step synthetic route shown in Scheme 1. The first step was the photochemical cyclodimerization of 9*H*-vinylcarbazole according to Ref. [14] followed by a *Vilsmeier* reaction to obtain the



Scheme 1



Scheme 2

mono- and dialdehydes **3a**,**b**. The last step was the condensation of **3a**,**b** with hydrazine derivatives with different substituents at the hydrazine N atom. The monohydrazone **4** and the compounds **5a**,**b** containing two hydrazine moieties with different substituents were prepared analogously.

CHC based dihydrazones were synthesized by the synthetic route shown in Scheme 2. The educt, 9-(6-(9*H*-carbazol-9-yl)-hexyl)-9*H*-carbazole (**7**), was prepared by the procedure reported in Ref. [15]. The second step was a *Vilsmeier* formylation followed by condensation of the resulting dialdehyde **8** with hydrazine derivatives to get dihydrazone compounds **9a**,**b** with different substituents at the hydrazone N atom.

The chemical structures of the new compounds were confirmed by IR, UV, ^{1}H NMR, and mass spectrometry (see Experimental). Their thermal properties were examined by thermogravimetric analysis (TGA) and differential scanning calorimetry (DSC). The thermal characteristics of the hydrazones are presented in Table 1.

All synthesized molecules are thermally stable; they decompose above 250°C. The *CCBC* based compounds showed higher thermal stability than the *CHC* based hydrazones, with **5b** exhibiting the highest thermal stability. Its 5% weight loss temperature ($T_{\text{dec-5\%}}$) is 300°C. The dihydrazone **9b**, of similar chemical structure but containing a hexyl spacer between the two carbazole moieties, showed the lowest thermal stability ($T_{\text{dec-5\%}} = 250$°C). Mono- and dihydrazones behave thermally very similar. Two stages are characteristic of the thermal degradation of

Table 1. Thermal characteristics of hydrazones

	T_g/°C	T_m/°C	$T_{dec\text{-}5\%}$/°C
4	88	195 (1st heating)	260
5a	136	–	265
5b	119	–	300
9a	91	–	260
9b	81	235 (1st heating)	250

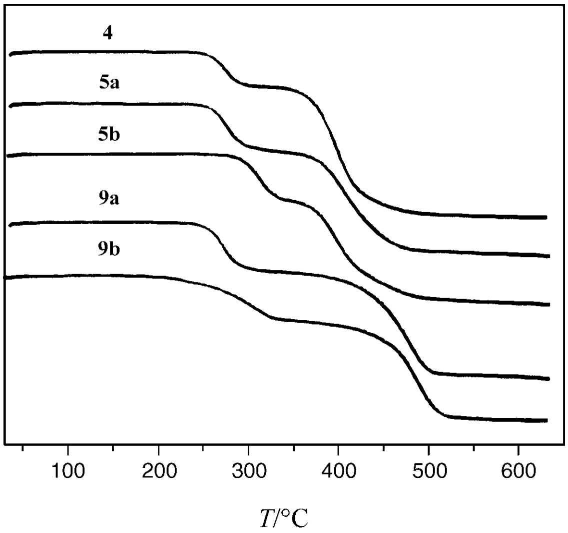
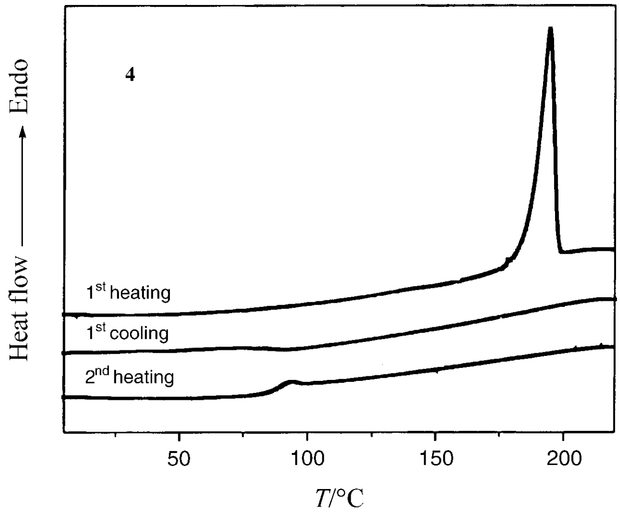


Fig. 1. TGA curves of hydrazones (heating rate 10°C/min, N_2 atmosphere)

all hydrazones (Fig. 1): the weight loss percentage at the first stage corresponds to the hydrazine moiety in the hydrazone molecules (apparently, the first step of thermal degradation of hydrazone molecules is due to the breaking of the azo bond); in the second step, decomposition of the products left after the first stage occurs.

The hydrazones **4**, **5a**,**b**, and **9a**,**b** formed stable glasses; **4** and **9b** were isolated after recrystallization. The first DSC heating runs of these compounds revealed melting peaks with maxima at 195 and 235°C. Recooling at a cooling rate of 10°C/min revealed no crystallization peaks, and only a glass transition was observed in the second DSC heating run. DSC curves of **4** are shown in Fig. 2a as an example.

The compounds **5a**,**b** and **9a** were isolated from column chromatography as amorphous powders. These compounds showed the glass transition already in the first DSC heating run, and no signal of crystallization in the cooling run or in the second heating run was observed. DSC curves of **5a** are shown in Fig. 2b. The DSC curve of the 1st heating run shows an enthalpy relaxation characteristic for a glass transition. The peak observed due to enthalpy relaxation is markedly reduced in the DSC curve of the 2nd heating performed after recooling the sample from the liquid state. The same holds for the glass transition of **5b**.



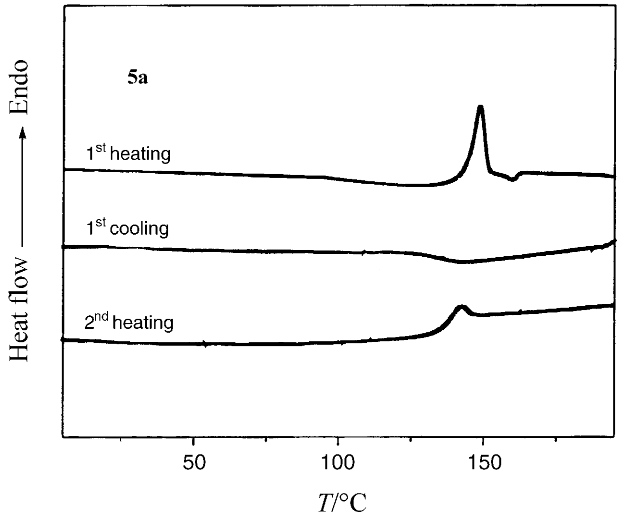


Fig. 2. DSC curves of hydrazones **4** and **5a** (heating/cooling rate 10°C/min, N_2 atmosphere)

The glass transition temperatures (T_g) of the molecular glasses are given in Table 1. The hydrazones containing the rigid *CCBC* moiety exhibited a *ca.* 40°C higher T_g than the *CHC* based hydrazones. The T_g values of **5b** and **9b** were by *ca.* 10°C lower than those of **5a** and **9a**. The comparison of the DSC data of mono- and dihydrazones **4** and **5a,b** showed that the introduction of the second hydrazine moiety into the molecules which led to the increase of their molar mass also increases the T_g values of the compounds.

All hydrazones absorb light in the region of 230–430 nm. The absorption spectra indicate that the hydrazones are strongly π-conjugated through the lone electron pair of the nitrogen atom and that the π-electrons are delocalized. As an example, the UV/Vis absorption spectra of *CCBC* based hydrazones **4** and **5a** are given in Fig. 3. For comparison, the UV/Vis spectrum of *CCBC* is also shown. It is evident that the introduction of the hydrazone moiety into the molecules of *CCBC*

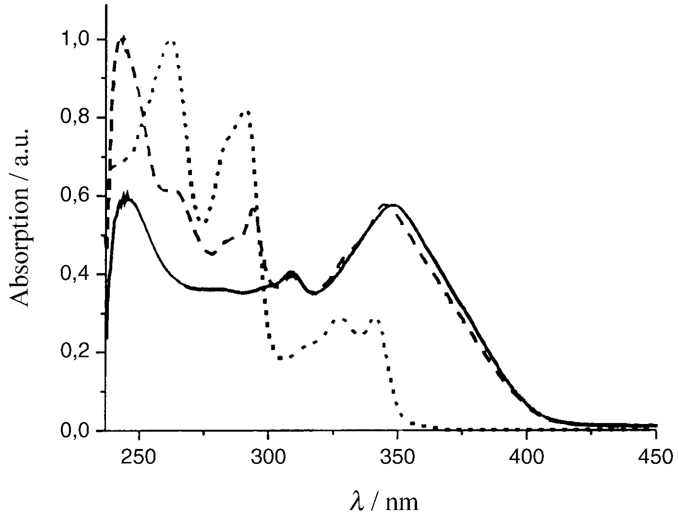


Fig. 3. Absorption spectra of dilute $CHCl_3$ solutions (10^{-5} *M*) of *CCBC* based hydrazones **4** (- - -) and **5a** (—) and *CCBC*

led to a marked bathochromic shift. The comparison of UV/Vis spectra of compounds containing one (**4**) and two hydrazone moieties (**5a**) showed that the introduction of the second hydrazone moiety did not lead to a marked bathochromic shift. Only a negligible bathochromic shift was observed for the methylphenyl substituted hydrazones **5a** and **9a** in comparison with the diphenyl substituted hydrazones **5b** and **9b**.

The hydrazones reported in this presentation are soluble in common organic solvents such as chloroform, *THF*, *etc*. Transparent films on substrates were prepared by the casting technique. The photoelectric properties of the hydrazones were preliminarily studied using such films. The ionization potential established by the photoemission technique ranged from 5.46 to 5.53 eV. The charge mobilities were measured by the time-of-flight technique. The highest hole drift mobility was observed in the amorphous film of **4**; it amounts to $7.6 \cdot 10^{-4}\,cm^2/Vs$ at an electric field of $9.3 \cdot 10^5\,V/cm$ at 22°C. The details of the photoelectric properties of these compounds will be reported in a forthcoming publication [16].

Experimental

^1H NMR spectra were obtained using a Bruker AC 250 NMR spectrometer (250 MHz). IR and UV/Vis spectra were recorded with Bio-Rad Digilab FTS-40 and Hitachi U-3000 spectrophotometers. Mass spectra were run on a Finnigan MAT 8500 mass spectrometer (70 eV). DSC measurements were carried out with a Perkin-Elmer DSC-7 instrument at 10°C/min heating/cooling rate under an N_2 atmosphere. TGA was performed on a Netzsch STA 409 apparatus with a data acquisition system 414/1 at 10°C/min heating rate under N_2. The ionization potentials were measured by the electron photoemission in air-method as described elsewhere [17]. The charge carrier mobilities were measured by the time of flight technique [18].

All chemicals were purchased from Aldrich and used as received without further purification, except 9*H*-vinylcarbazole (**1**) which was recrystallized from EtOH before use. All organic solvents were purified and dried as usual.

9-(2-(9H-Carbazol-9-yl)-cyclobutyl)-9H-carbazole (CCBC) (**2**; $C_{28}H_{22}N_2$)

A solution of 24 g **1** in 250 cm^3 acetone was irradiated (Hg lamp DK$_s$Sh-1000, $\lambda = 245$–1045 nm) for 30 h at 25°C. Air was bubbled through the solution continuously. The precipitated product was filtered and recrystallized from an acetone ethanol = 1:1.

Yield: 17.5 g (73%); m.p.: 195–196.5°C (Ref. [19]: 192°C); IR (KBr): $\nu = 3050, 3020$ (ar. C–H), 2963 (alk. C–H), 1597, 1483 (ar. C=C), 1271 (C–N) cm^{-1}; ^{1}H NMR ($CDCl_3$, δ, 250 MHz): 2.80 (m, CH_2 of cyclobutane), 3.10 (m, CH_2 of cyclobutane), 6.30 (m, 2H, CH of cyclobutane), 7.2 (m, 4H ar.), 7.42 (m, 4H ar.), 7.58 (d, $J = 7.8$ Hz, 4H ar.), 8.07 (d, $J = 8.0$ Hz, 4H ar.) ppm.

9-(2-(9H-Carbazol-9-yl)-cyclobutyl)-9H-carbazole-3-carbaldehyde (**3a**; $C_{29}H_{22}N_2O$)

To 0.84 cm^3 of dry *DMF* (10.8 mmol) 0.84 cm^3 $POCl_3$ (9 mmol) were added dropwise at 0°C under N_2. A solution of 2.317 g (6 mmol) **2** in 20 cm^3 dry *o*-dichlorobenzene was added stepwise. The reaction mixture was stirred at 85°C for 20 h. Then it was cooled to room temperature, poured in to ice-H_2O, and neutralized with sodium acetate. The aqueous solution was extracted several times with $CHCl_3$. The $CHCl_3$ solution was washed with H_2O, dried with anh. Na_2SO_4, filtered, and distilled. The crude product was purified by column chromatography (eluent: acetone:hexane = 1:8).

Yield: 1.12 g (45%); m.p.: 96–89°C (Ref. [20]: 89–92°C); IR (KBr): $\nu = 3056$ (ar. C–H), 2965 (alk. C–H), 1678 (CHO), 1597, 1480 (ar. C=C), 1280 (C–N) cm^{-1}; MS: $m/z = 414$ (M^+), 221, 193, 167; ^{1}H NMR ($CDCl_3$, δ, 250 MHz): 2.80 (m, CH_2 of cyclobutane), 3.10 (m, CH_2 of cyclobutane), 6.30 (m, 2H, CH of cyclobutane), 7.2–7.82 (m, 11H ar.), 7.90 (d, $J = 8.8$ Hz, 1H ar.), 8.10 (m, 2H ar.), 8.60 (s, 1H ar.), 10.05 (s, 1H of CHO) ppm.

9-(2-(3-Formyl-9H-carbazol-9-yl)-cyclobutyl)-9H-carbazole-3-carbaldehyde (**3b**; $C_{30}H_{22}N_2O_2$)

Compound **3b** was prepared by the same procedure as **3a**.

Yield: 88%; m.p.: 122–125°C (Ref. [20]: 118–120°C); IR (KBr): $\nu = 3050, 3013$ (ar. C–H), 2964 (alk. C–H), 1685 (CHO), 1594, 1489 (ar. C=C), 1280 (C–N) cm^{-1}; MS: $m/z = 442$ (M^+), 414, 221, 192, 166; ^{1}H NMR ($CDCl_3$, δ, 250 MHz): 2.90 (m, CH_2 of cyclobutane), 3.20 (m, CH_2 of cyclobutane), 6.30 (m, 2H, CH of cyclobutane), 7.2–7.82 (m, 11H ar.), 7.90 (d, $J = 7.5$ Hz, 1H ar.), 8.10 (m, 2H ar.), 8.60 (s, 1H ar.), 10.05 (s, 1H of CHO) ppm.

9-(2-(9H-Carbazol-9-yl)-cyclobutyl)-9H-carbazole-3-carbaldehyde N,N-diphenylhydrazone (**4**; $C_{41}H_{32}N_4$)

A solution of 1 g (4.8 mmol) N,N-diphenylhydrazine hydrochloride in 30 cm^3 EtOH was added dropwise to a solution of 0.5 g (1.2 mmol) **3a** in 30 cm^3 EtOH under stirring. The reaction mixture was refluxed until total consumption of **3a** (TLC control) and then cooled to room temperature. The precipitated product was filtered and purified by recrystallization from EtOH followed by column chromatography (eluent: $CHCl_3$).

Yield: 0.58 g (83%); m.p.: 186–188°C; IR (KBr): $\nu = 3053, 3022$ (ar. C–H), 2967 (alk. C–H), 1625 (C=N), 1591, 1492 (ar. C=C), 1271 (C–N) cm^{-1}; MS: $m/z = 580$ (M^+), 413, 387, 218, 193, 167; ^{1}H NMR ($CDCl_3$, δ, 250 MHz): 2.75 (m, CH_2 of cyclobutane), 3.10 (m, CH_2 of cyclobutane), 6.25 (s, 2H, CH of cyclobutane), 7.1–7.25 (m, 8H ar.), 7.28–7.6 (m, 13H ar.), 7.8 (d, $J = 10$ Hz, 1H ar.), 8.0 (m, 3H ar.), 8.15 (s, 1H of methine) ppm.

9-(2-(3-((2,2-Diphenylhydrazono)-methyl)-9H-carbazol-9-yl)-cyclobutyl)-9H-carbazole-3-carbaldehyde N,N-diphenylhydrazone (**5a**; $C_{54}H_{42}N_6$)

Compound **5a** was prepared by the same procedure as **4**.

Yield: 84%; IR (KBr): $\nu = 3058, 3023$ (ar. C–H), 2961, 2924 (alk. C–H), 1625 (C=N), 1590, 1492 (ar. C=C), 1271 (C–N) cm^{-1}; MS: $m/z = 605$, 438, 413, 385, 218, 192, 167, 77; ^{1}H NMR ($CDCl_3$, δ, 250 MHz): 2.76 (m, CH_2 of cyclobutane), 3.10 (m, CH_2 of cyclobutane), 6.23 (m, 2H, CH of cyclobutane), 7.10–7.25 (m, 14H ar.), 7.30–7.58 (m, 16H ar.), 7.8 (d, $J = 9.6$ Hz, 2H ar.), 8.0 (d, $J = 9.6$ Hz, 2H ar.), 8.14 (s, 2H of methine) ppm.

9-(2-(3-((2-Ethyl-2-phenylhydrazono)-methyl)-9H-carbazol-9-yl)-cyclobutyl)-9H-carbazole-3-carbaldehyde N-methyl-N-phenylhydrazone (**5b**; $C_{44}H_{38}N_6$)

Compound **5b** was prepared by the same procedure as **4** employing N-methyl-N-phenylhydrazine instead of N,N-diphenylhydrazine hydrochloride.

Yield: 73%; IR (KBr): $\nu = 3053, 3022$ (ar. C–H), 2967 (alk. C–H), 1625 (C=N), 1591, 1492 (ar. C=C), 1271 (C–N) cm^{-1}; MS: $m/z = 605$, 351, 218, 192, 165, 106, 77; ^{1}H NMR ($CDCl_3$, δ, 250 MHz): 2.77 (m, CH_2 of cyclobutane), 3.10 (m, CH_2 of cyclobutane), 3.45 (s, 6H of methyl), 6.26 (s, 2H, CH of cyclobutane), 6.86–6.97 (m, 2H ar.), 7.15–7.25 (m, 2H ar.), 7.27–7.45 (m, 10H ar.), 7.46–7.6 (m, 4H ar.), 7.68 (s, 2H ar.), 7.84 (d, $J = 9.6$ Hz, 2H ar.), 8.10 (d, $J = 9.6$ Hz, 2H ar.), 8.29 (s, 2H of methine) ppm.

9-(6-(9H-Carbazol-9-yl)-hexyl)-9H-carbazole (**7**; $C_{30}H_{28}N_2$)

20 g (119.7 mmol) **6** and 4.6 cm^3 (29.9 mmol) 1,6-dibromohexane were dissolved in 80 cm^3 acetone. Then, 0.4 g (1.2 mmol) tetrabutylammoniumsulfate and, in portions, 3.4 g (59.9 mmol) KOH were added. After refluxing for 72 h, the solvent was removed by distillation. The residue was dissolved in diethyl ether, and the insoluble part was filtered. Diethyl ether was removed by distillation, and the crude product was recrystallized from acetone.

Yield: 5.4 g (43%); m.p.: 126–128°C (Ref. [21]: 127.7°C). IR (KBr): $\nu = 3060$ (ar. C–H), 2950 (alk. C–H), 1590, 1490 (ar. C=C), 1230 (C–N) cm^{-1}; MS: $m/z = 416$ (M^+), 180; ^{1}H NMR ($CDCl_3$, δ, 250 MHz): 1.25–1.45 (m, 4H alk.), 1.70–1.90 (m, 4H alk.), 4.23 (m, 4H alk.), 7.20 (m, 4H ar), 7.20 (d, $J = 8.1$ Hz, 4H ar), 7.43 (m, 4H ar), 8.08 (d, $J = 8.4$ Hz, 4H ar) ppm.

9-(6-(3-Formyl-9H-carbazol-9-yl)-hexyl)-9H-carbazole-3-carbaldehyde (**8**; $C_{32}H_{28}N_2O_2$)

Compound **8** was synthesized by the same procedure as **3a**.

Yield: 86%; m.p.: 161–163°C (Ref. [21]: 160°C); IR (KBr): $\nu = 3060$ (ar. C–H), 2950 (alk. C–H), 1683 (CHO), 1590, 1490 (ar. C=C), 1230 (C–N) cm^{-1}; MS: $m/z = 472$ (M^+), 444, 208, 180; ^{1}H NMR ($CDCl_3$, δ, 250 MHz): 1.20–1.50 (m, 4H alk.), 1.70–1.95 (m, 4H alk.), 4.30 (m, 4H alk.), 7.20–7.40 (m, 6H ar), 7.45–7.60 (m, 2H ar), 7.93 (d, $J = 8.1$ Hz, 2H ar), 8.18 (d, $J = 9.0$ Hz, 2H ar), 8.60 (s, 2H ar), 10.00 (s, 2H of CHO) ppm.

9-(6-(3-((-2,2-Diphenylhydrazono)-methyl)-9H-carbazol-9-yl)-hexyl)-9H-carbazole-3-carbaldehyde N,N-diphenylhydrazone (**9a**; $C_{56}H_{48}N_6$)

Compound **9a** was synthesized by the same procedure as **4**. After 24 h, the hot reaction mixture was poured into cold isopropanol. The precipitated product was filtered and washed with isopropanol. The crude product was purified by column chromatography (eluent: hexane:$CHCl_3 = 3:1$).

Yield: 46%; IR (KBr): $\nu = 3060$ (ar. C–H), 2950 (alk. C–H), 1630 (C=N), 1590, 1490 (ar. C=C), 1230 (C–N) cm^{-1}; MS: $m/z = 804$ (M^+), 635, 466, 169; ^{1}H NMR ($CDCl_3$, δ, 250 MHz): 1.25–1.45 (m, 4H alk.), 1.70–1.90 (m, 4H alk.), 4.20 (m, 4H alk.), 7.10–7.35 (m, 20H ar), 7.35–7.55 (m, 10H ar), 7.83 (d, $J = 6.3$ Hz, 2H ar), 8.08 (d, $J = 6.3$ Hz, 2H ar), 8.20 (s, 2H of methine) ppm.

9-(6-(3-((-2-Methyl-2-phenylhydrazono)-methyl)-9H-carbazol-9-yl)-hexyl)-9H-carbazole-3-carbaldehyde N-methyl-N-phenylhydrazone (**9b**; $C_{46}H_{44}N_6$)

Compound **9b** was synthesized by the same procedure as **4**. After 48 h, the hot reaction mixture was poured into cold isopropanol. The precipitated product was filtered and washed with isopropanol. The crude product was recrystallized from $CHCl_3$:PrOH = 4:1.

Yield: 63%; m.p.: 224–226°C; IR (KBr): $\nu = 3050$ (ar. C–H), 2940 (alk. C–H), 1630 (C=N), 1590, 1490 (ar. C=C), 1240 (C–N) cm^{-1}; MS: $m/z = 680$ (M^+), 573, 469, 180, 106; ^1H NMR ($CDCl_3$, δ, 250 MHz): 1.30–1.45 (m, 4H alk.), 1.70–1.90 (m, 4H alk.), 3.45 (s, 6H methyl), 4.23 (m, 4H alk.), 6.93 (m, 2H ar), 7.15–7.50 (m, 16H ar), 7.70 (s, 2H ar.), 7.88 (d, $J = 6.3$ Hz, 2H ar), 8.13 (d, $J = 6.3$ Hz, 2H ar), 8.35 (s, 2H of methine) ppm.

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