Electronic Supplementary Material (ESI) for New Journal of Chemistry.

Synthesis, optical and electrochemical properties of a series of push-pull dyes based on the 4-(9-ethyl-9*H*-carbazol-3-yl)-4-phenylbuta-1,3-dienyl donor

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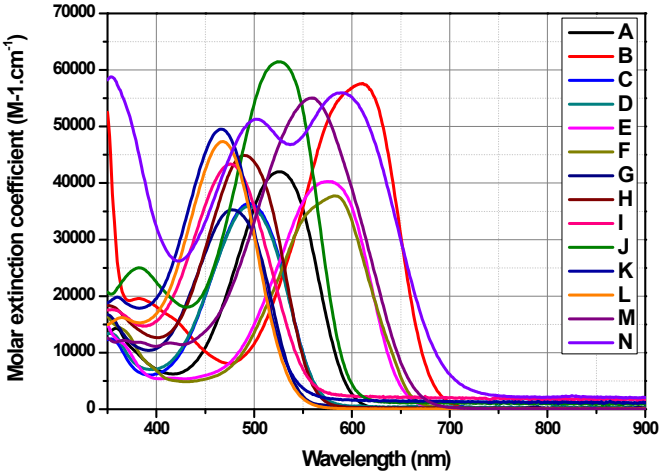
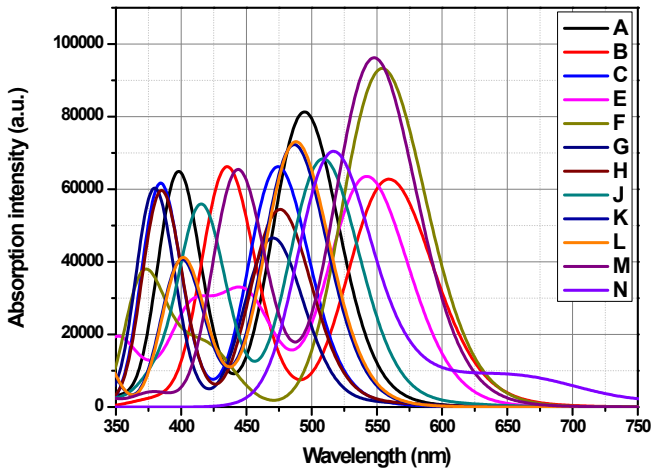
Comparison of the experimental UV spectra in chloroform (SI 1)

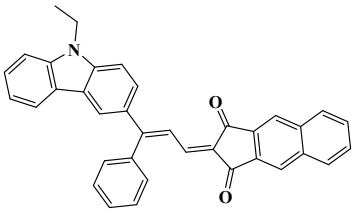


Comparison of all simulated UV spectra in dichloromethane (SI 2)

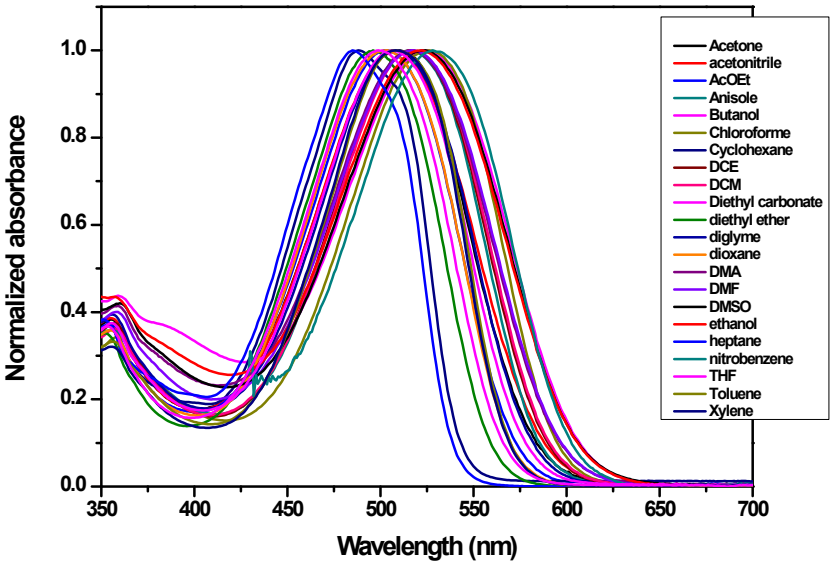


UV-visible absorption spectra of compounds A-N in 23 different solvents

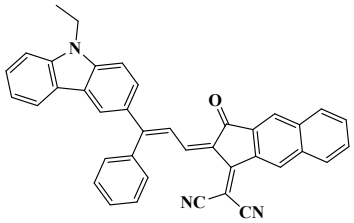
Compound A



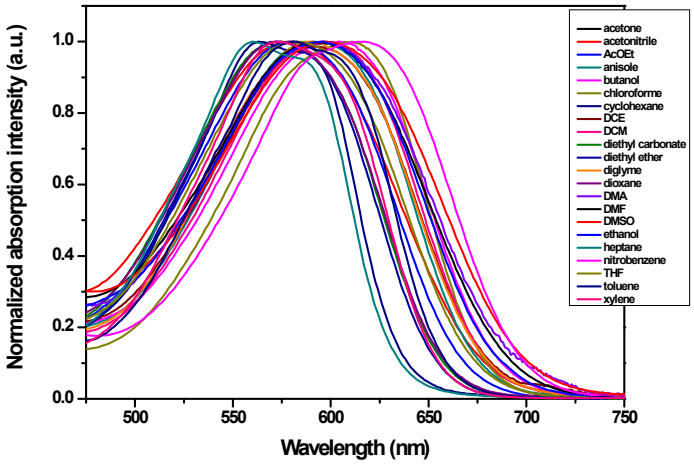
Chemical Formula: $C_{36}H_{25}NO_2$
Molecular Weight: 503.6010



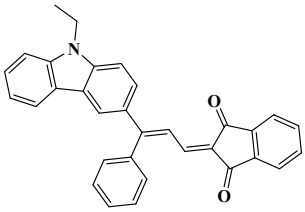
Compound B



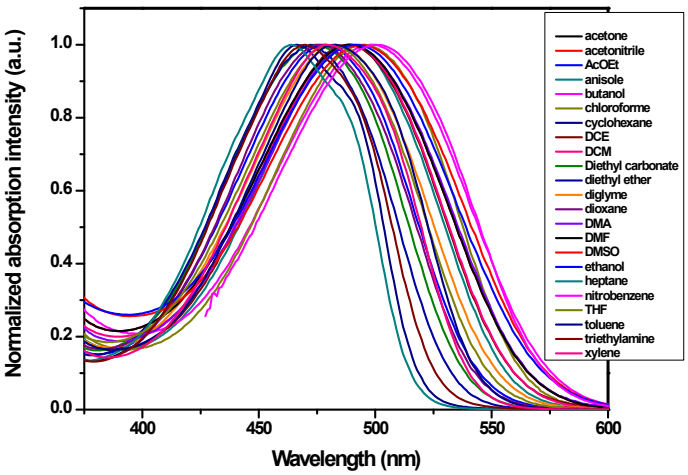
Chemical Formula: $C_{39}H_{25}N_3O$
Molecular Weight: 551.6490



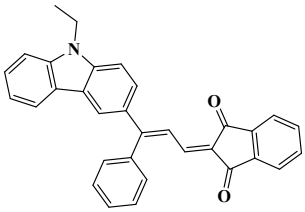
Compound C



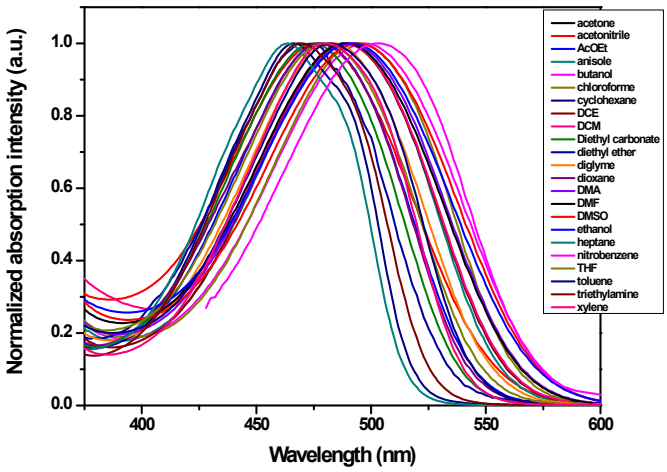
Chemical Formula: $C_{32}H_{23}NO_2$
Molecular Weight: 453.5410



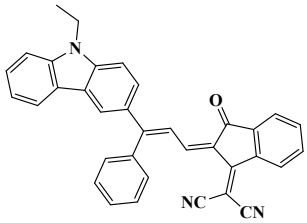
Compound D



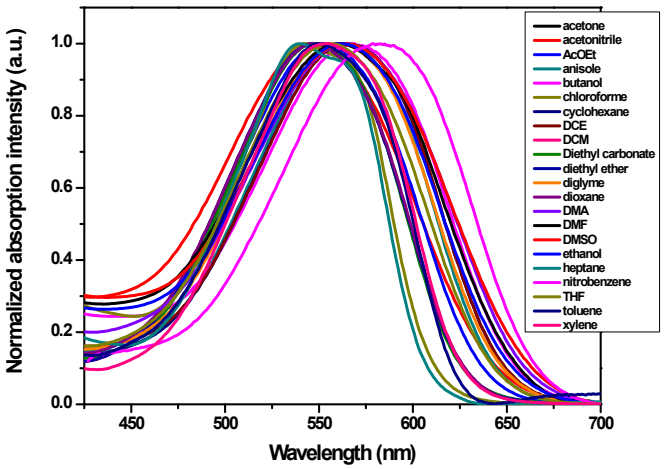
Chemical Formula: $C_{32}H_{23}NO_2$
Molecular Weight: 453.5410



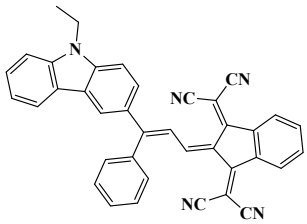
Compound E



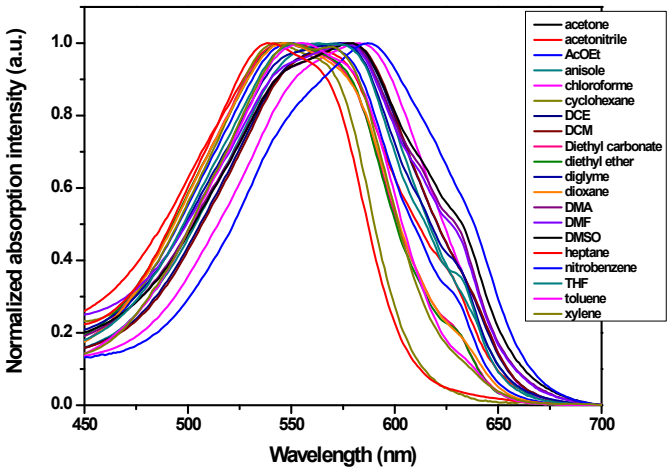
Chemical Formula: $C_{35}H_{23}N_3O$
Molecular Weight: 501.5890



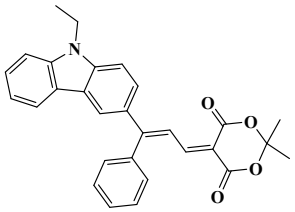
Compound F



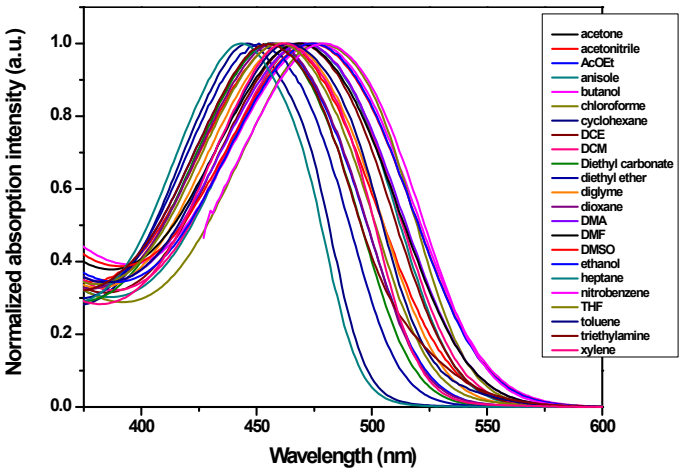
Chemical Formula: $C_{38}H_{23}N_5$
Molecular Weight: 549.6370



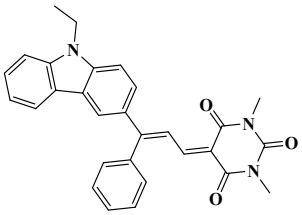
Compound G



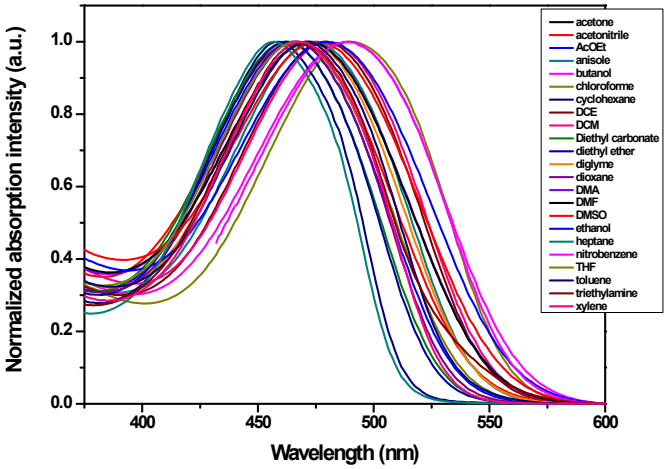
Chemical Formula: $C_{29}H_{25}NO_4$
Molecular Weight: 451.5220



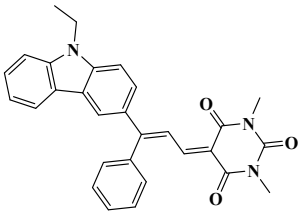
Compound H (mixture of isomers)



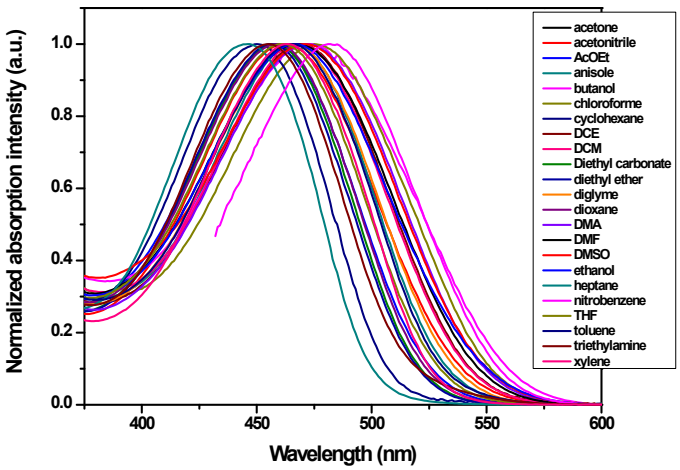
Chemical Formula: $C_{29}H_{25}N_3O_3$
Molecular Weight: 463.5370



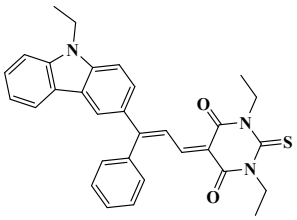
Compound I (pure isomer)



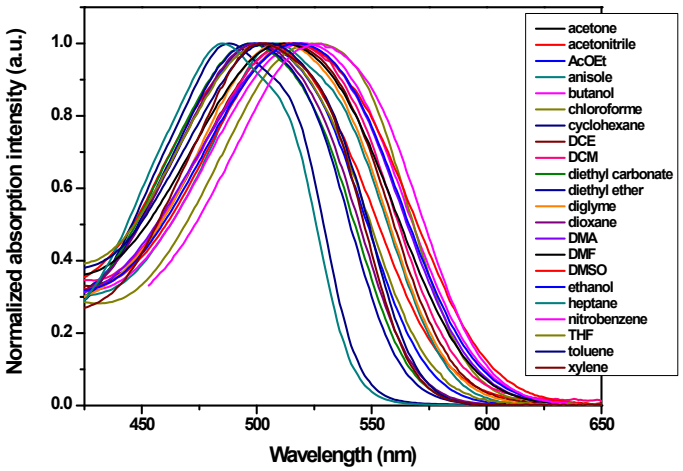
Chemical Formula: $C_{29}H_{25}N_3O_3$
Molecular Weight: 463.5370



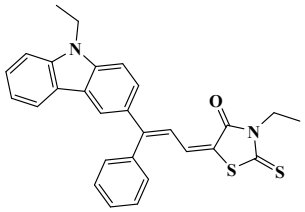
Compound J



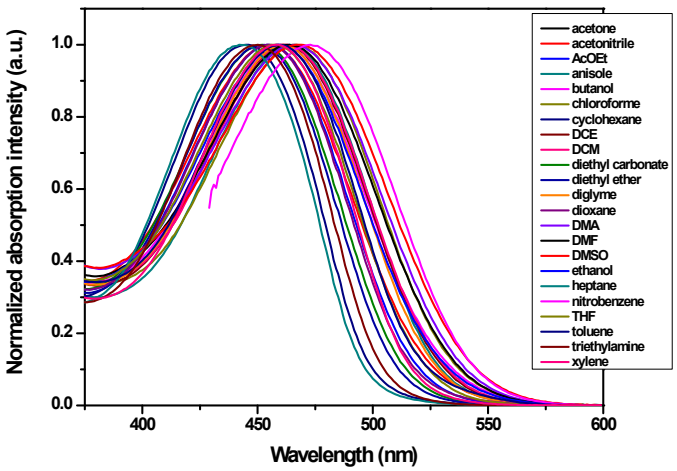
Chemical Formula: $C_{31}H_{29}N_3O_2S$
Molecular Weight: 507.6520



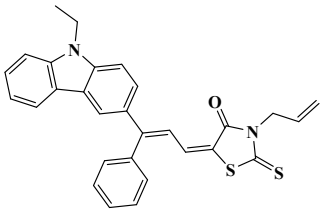
Compound K



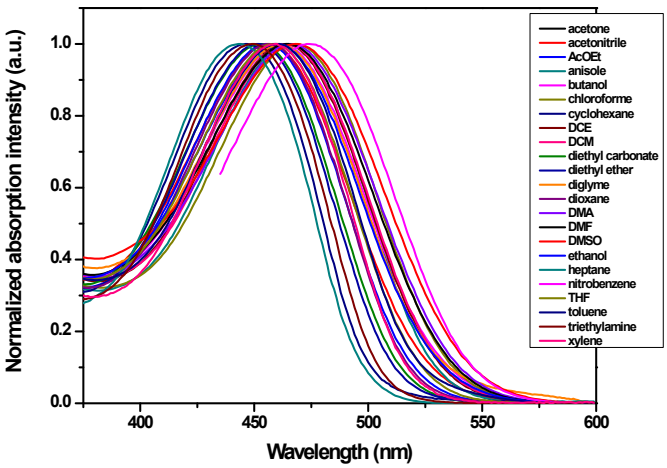
Chemical Formula: $C_{28}H_{24}N_2OS_2$
Molecular Weight: 468.6330



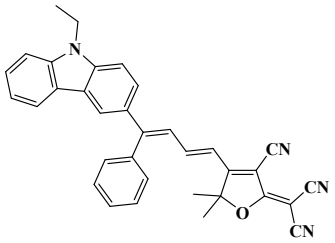
Compound L



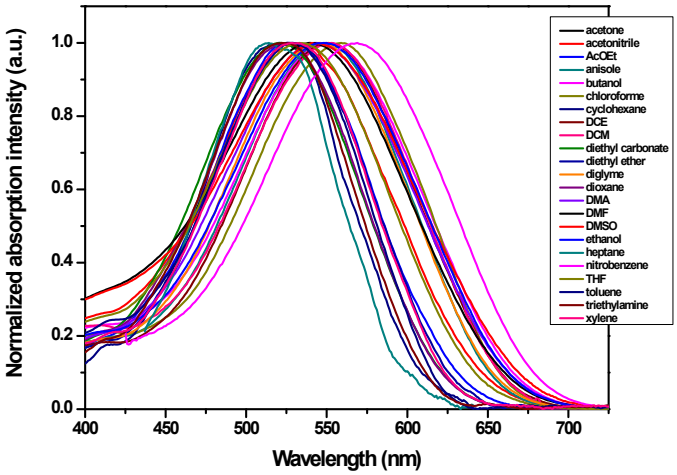
Chemical Formula: $C_{29}H_{24}N_2OS_2$
Molecular Weight: 480.6440



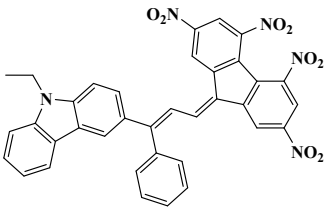
Compound M



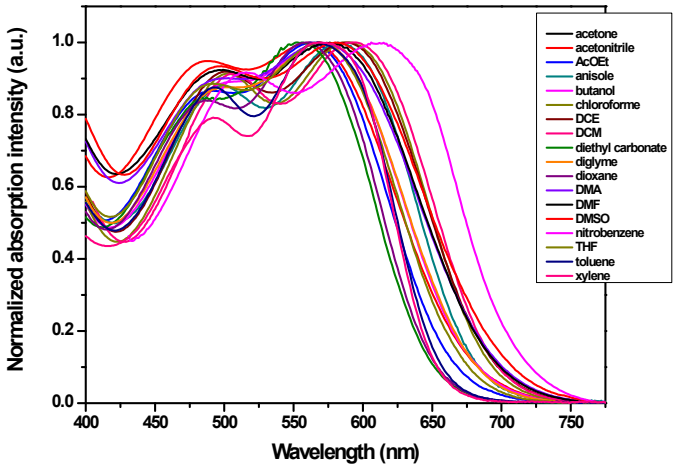
Chemical Formula: $C_{34}H_{26}N_4O$
Molecular Weight: 506.6090



Compound N

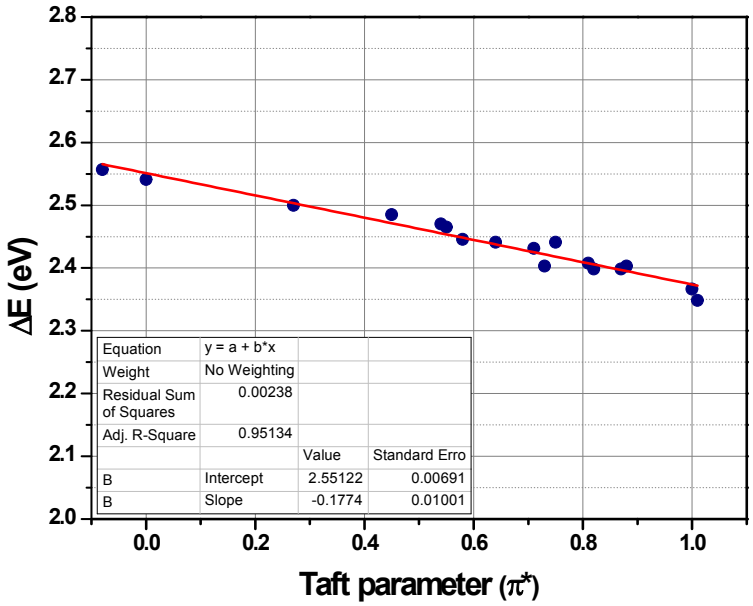


Chemical Formula: $C_{36}H_{23}N_5O_8$
Molecular Weight: 653.6070



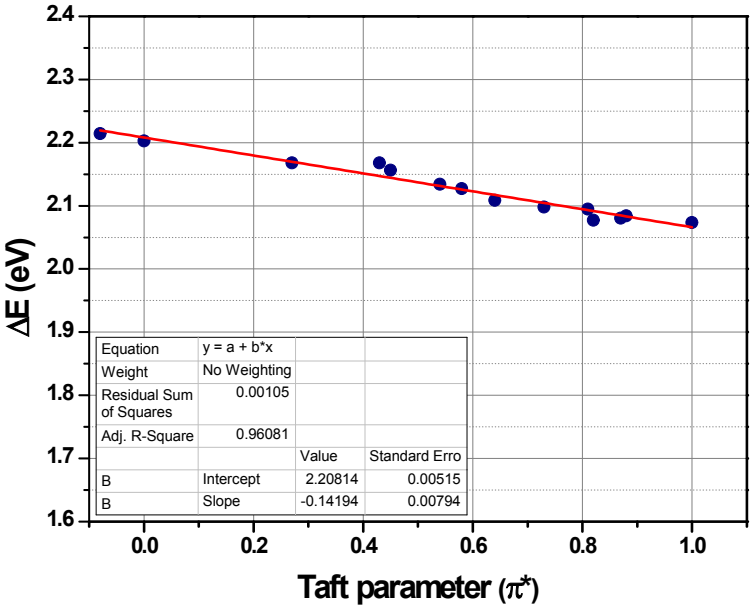
Position of the absorption maxima of compounds A-N in 23 solvents of different polarities vs. the Kamlet–Taft parameters π*

Compound A



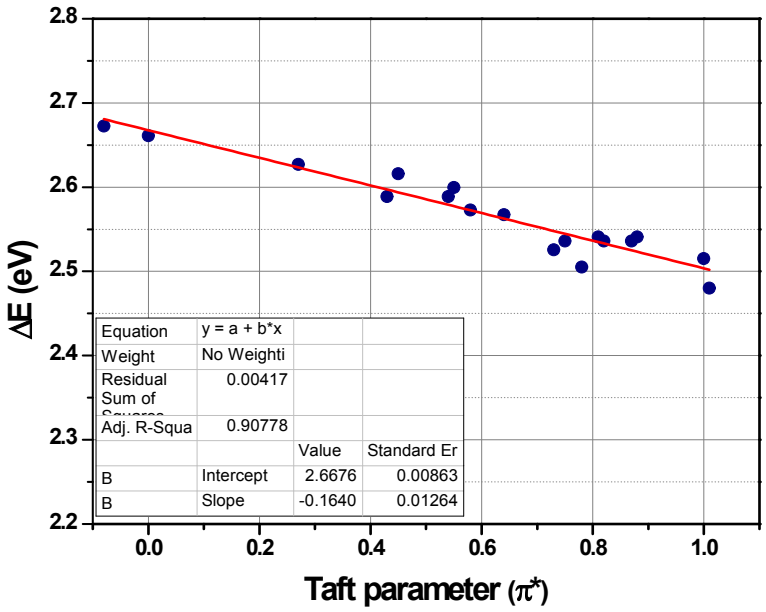
$y = -0.1774\ x + 2.5512$ $R^2 = 0.951$

Compound B



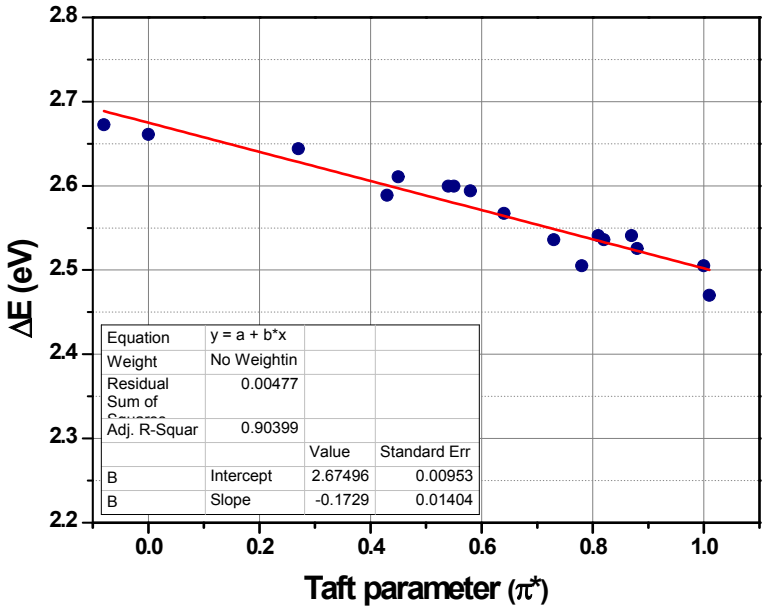
$y = -0.14194\ x + 2.20814$ $R^2 = 0.961$

Compound C



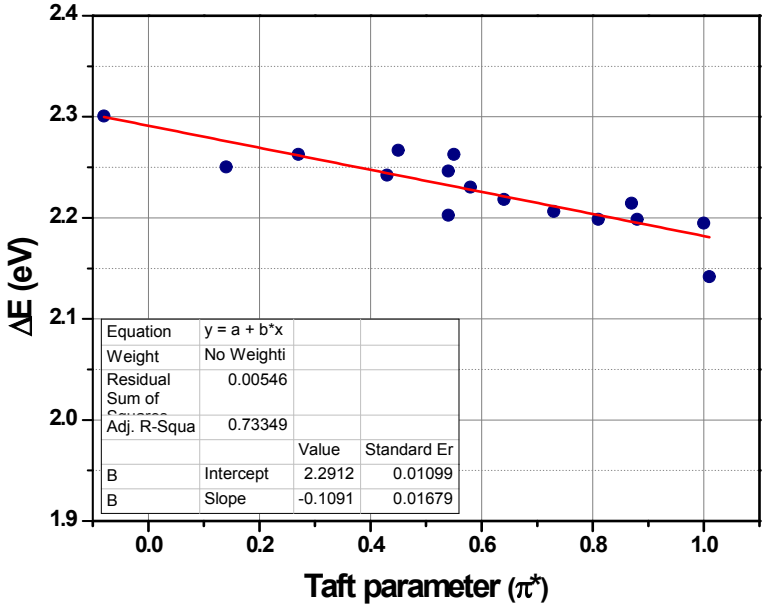
$y = -0.1640\ x + 2.6676$ $R^2 = 0.908$

Compound D



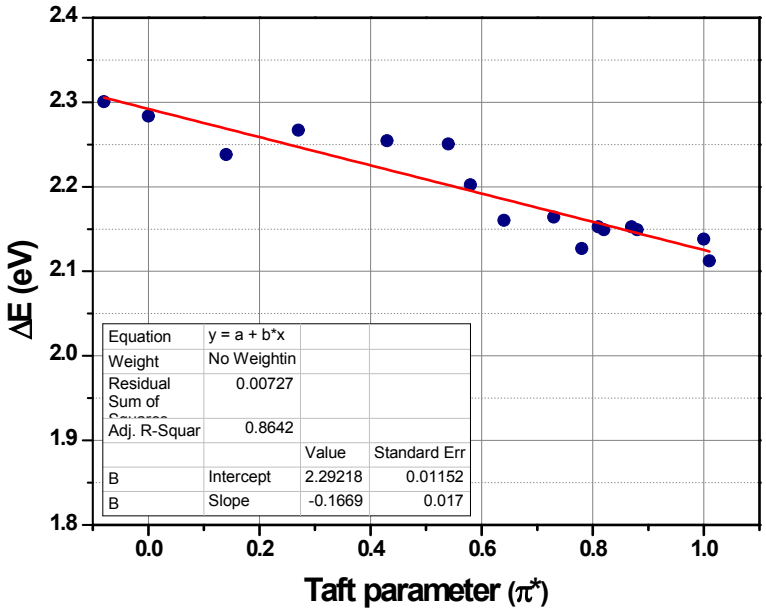
$y = -0.1729\ x + 2.67496$ $R^2 = 0.903$

Compound E



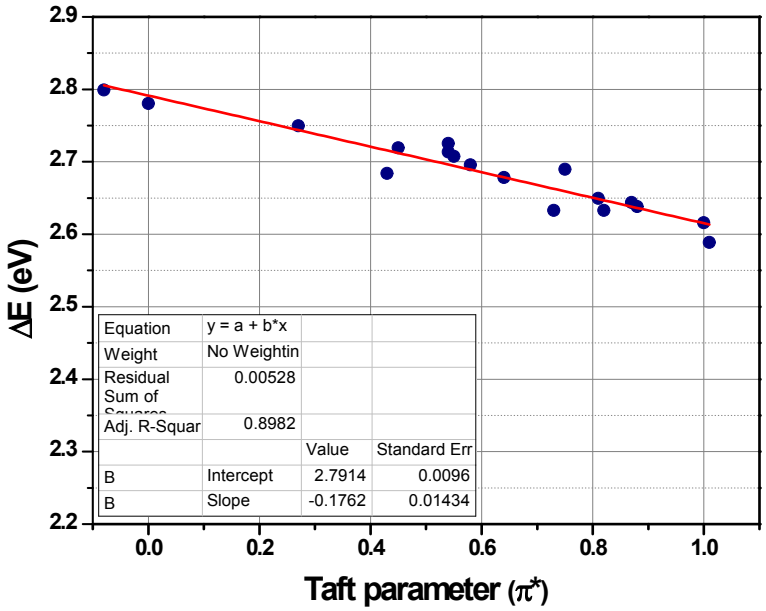
$y = -0.1091\ x + 2.2912$ $R^2 = 0.733$

Compound F



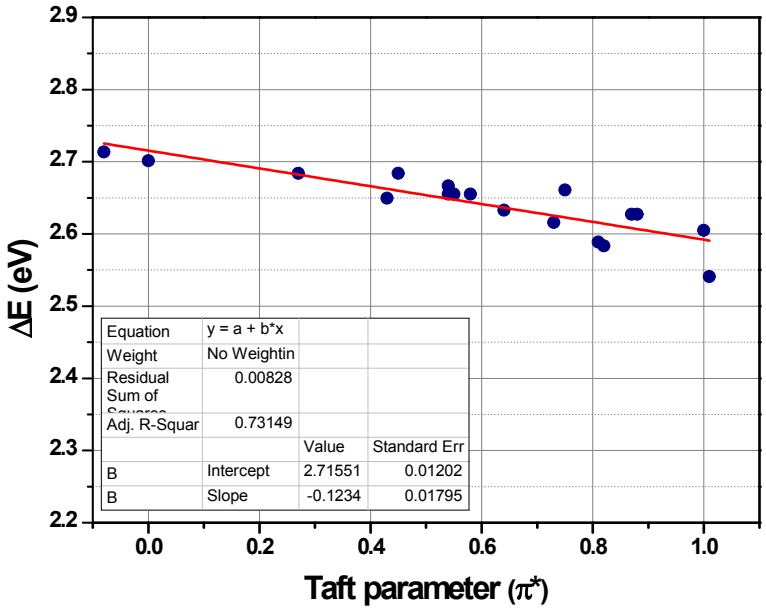
$y = -0.1669\ x + 2.29218$ $R^2 = 0.864$

Compound G



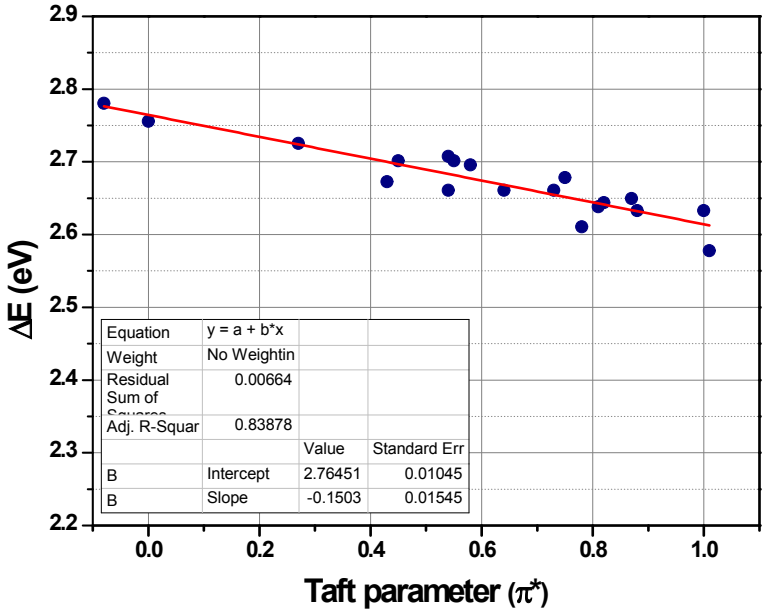
$y = -0.1762\ x + 2.7914$ $R^2 = 0.898$

Compound H



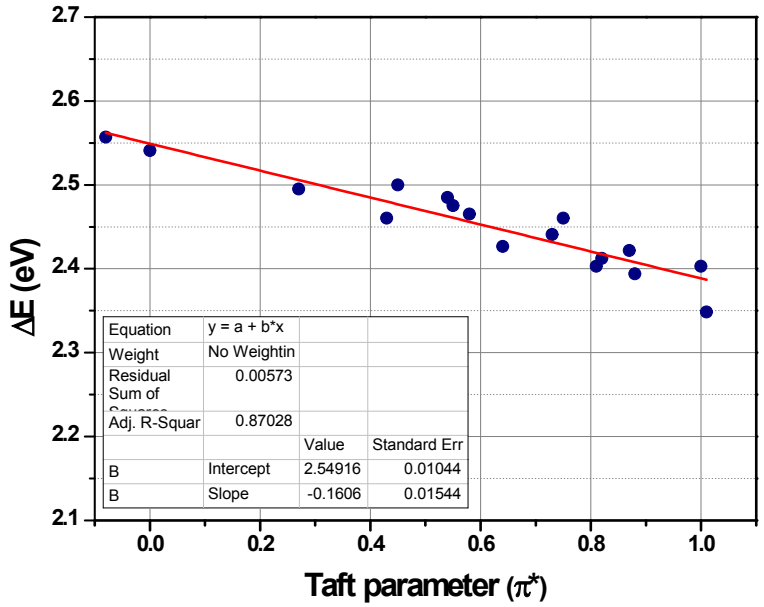
$y = -0.1234\ x + 2.71551$ $R^2 = 0.731$

Compound I



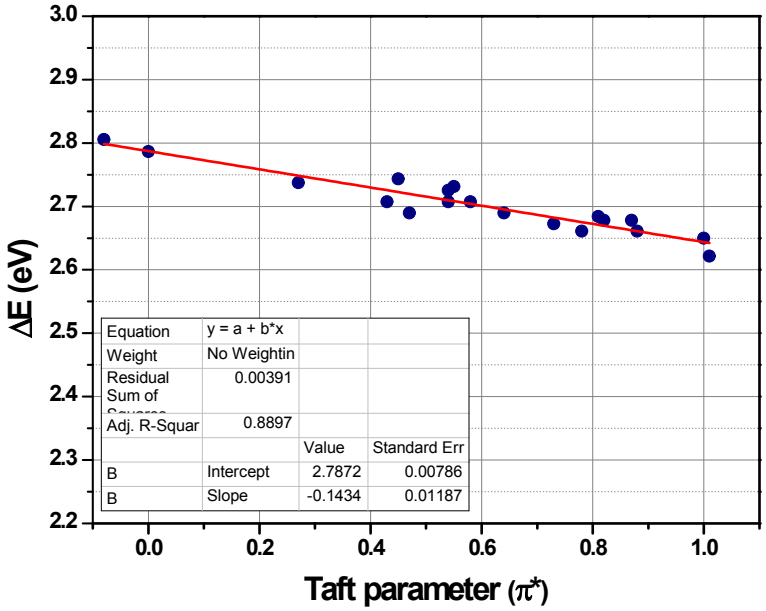
$y = -0.1503\ x + 2.76451$ $R^2 = 0.838$

Compound J



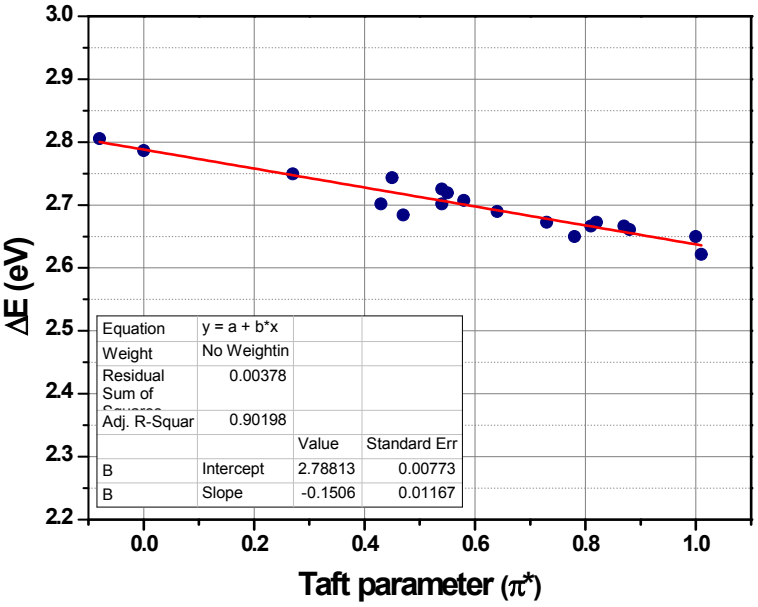
$y = -0.1606\ x + 2.54916$ $R^2 = 0.871$

Compound K



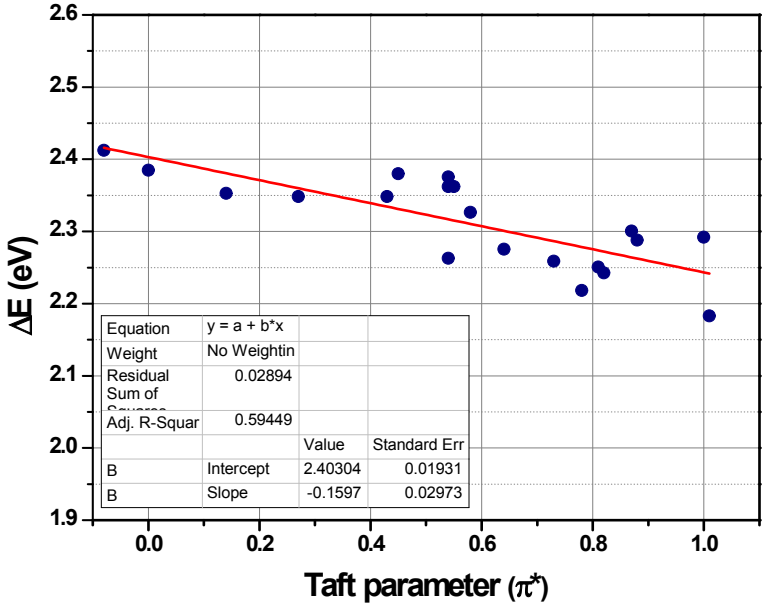
$y = -0.1434\ x + 2.7872$ $R^2 = 0.890$

Compound L



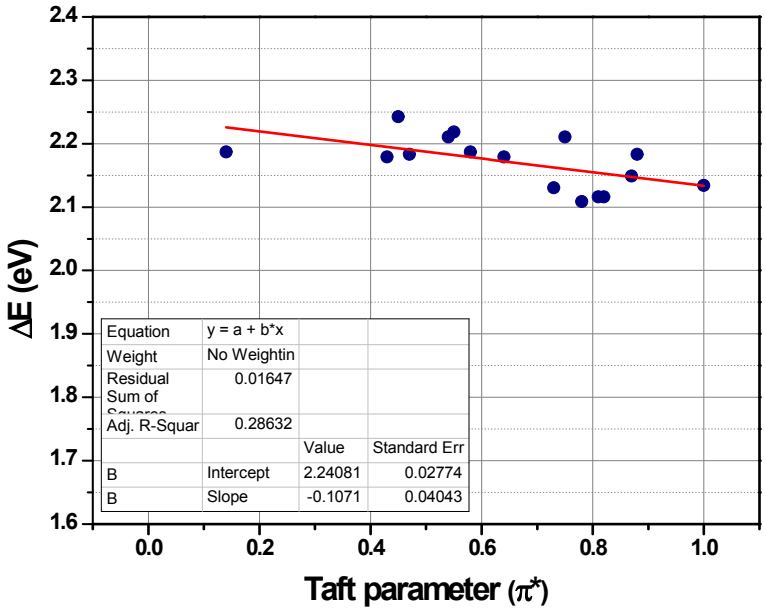
$y = -0.1506\ x + 2.78813$ $R^2 = 0.902$

Compound M



$y = -0.1597\ x + 2.40304$ $R^2 = 0.594$

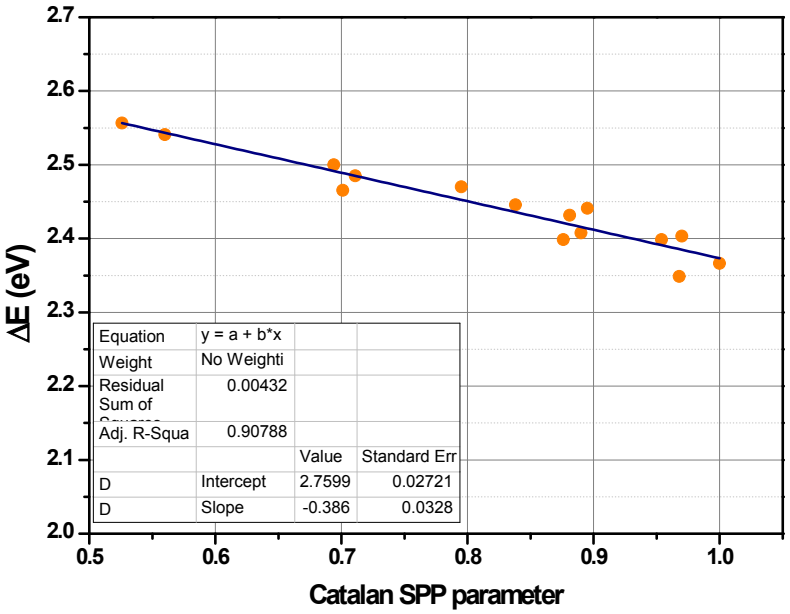
Compound N



$y = -0.1071\ x + 2.24081$ $R^2 = 0.286$

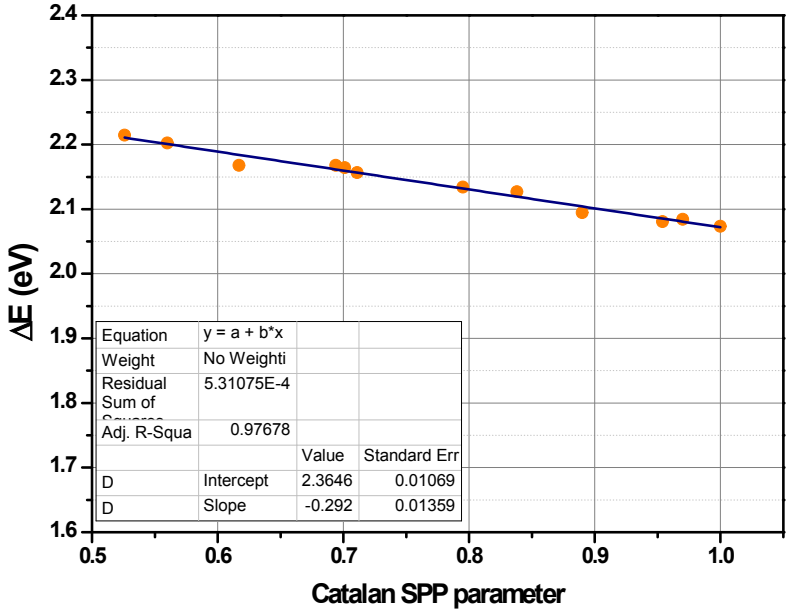
Position of the absorption maxima of A-N in 23 solvents of different polarities vs. the Catalan solvent polarity/polarizability (SPP) scale

Compound A



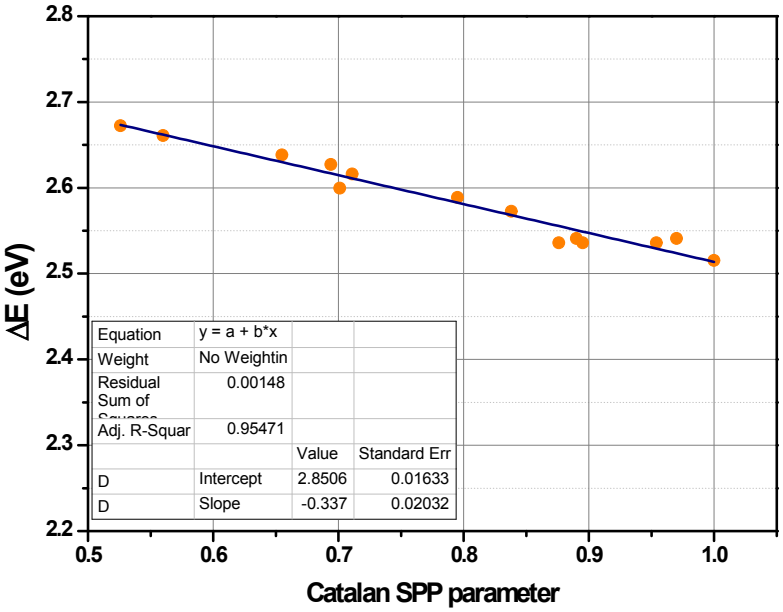
$y = -0.386\ x + 2.7599$ $R^2 = 0.908$

Compound B



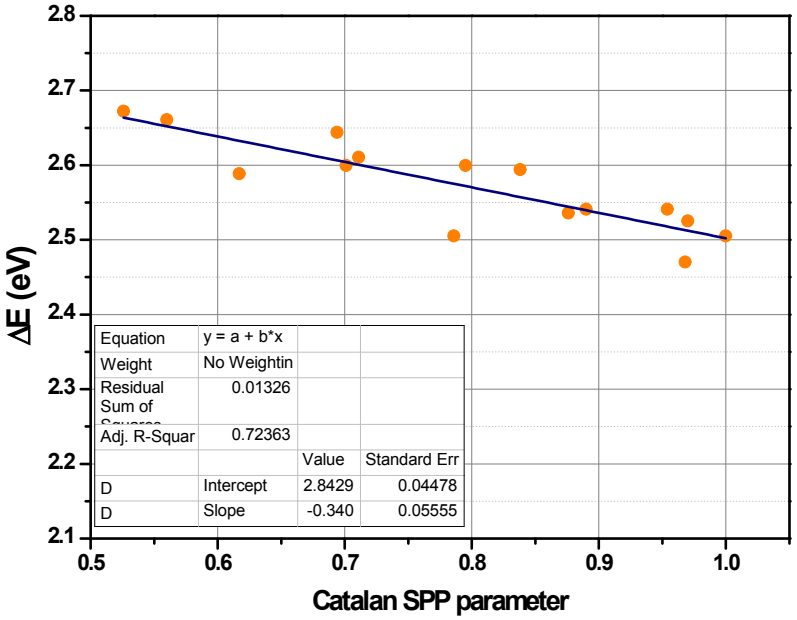
$y = -0.292\ x + 2.3646$ $R^2 = 0.977$

Compound C



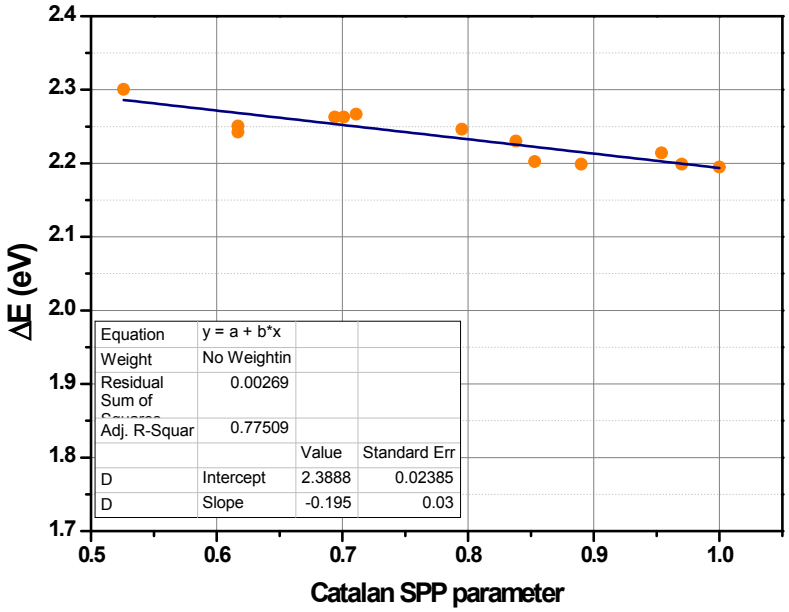
$y = -0.337\ x + 2.8506$ $R^2 = 0.955$

Compound D



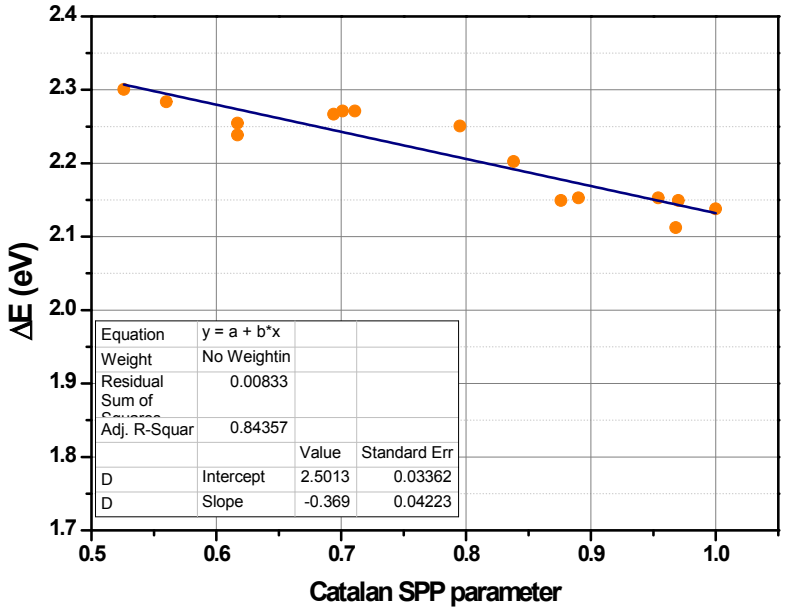
$y = -0.340\ x + 2.8429$ $R^2 = 0.723$

Compound E



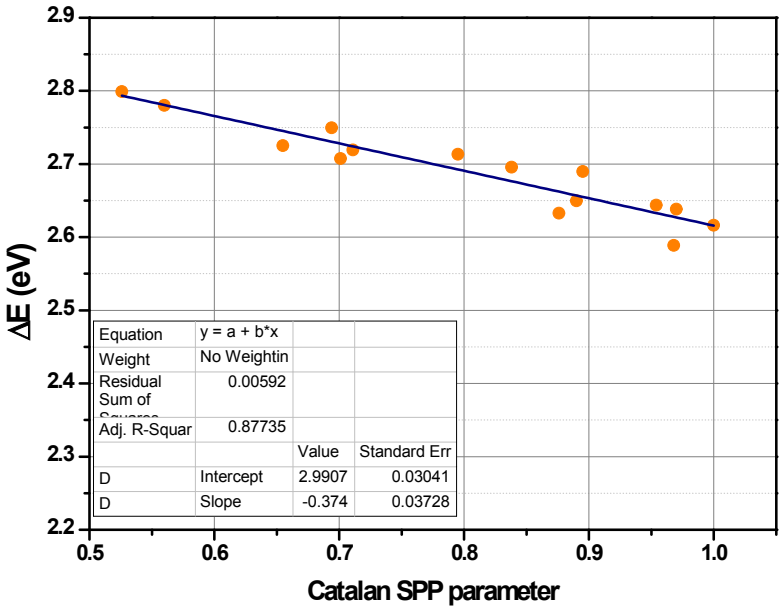
$y = -0.195\ x + 2.3888$ $R^2 = 0.775$

Compound F



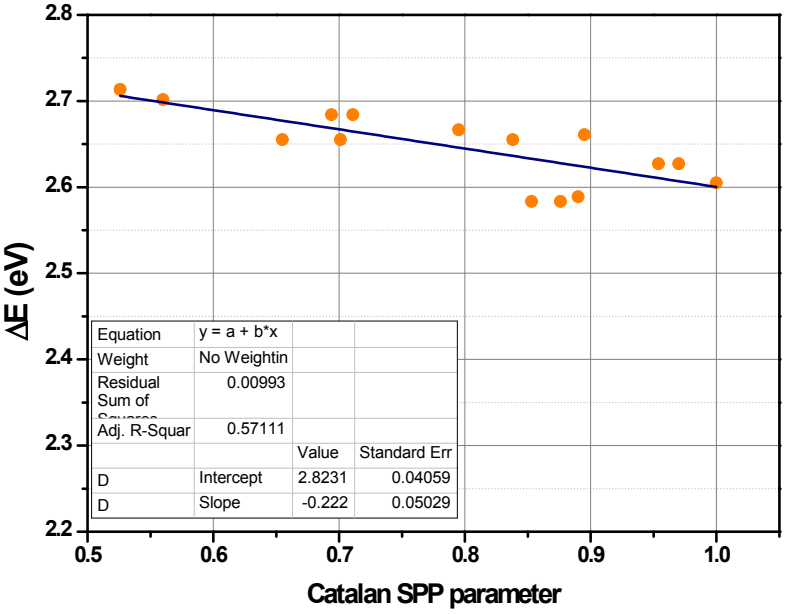
$y = -0.369\ x + 2.5013$ $R^2 = 0.843$

Compound G



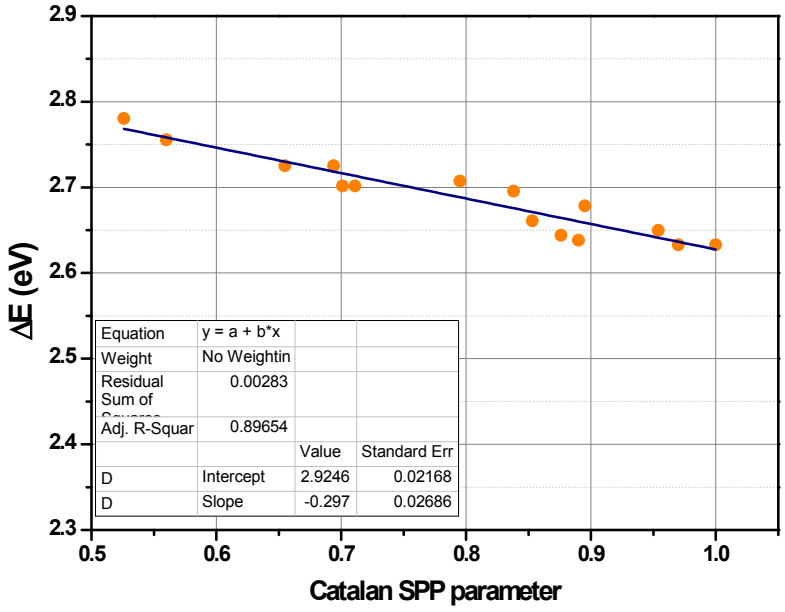
$y = -0.374\ x + 2.9907$ $R^2 = 0.877$

Compound H



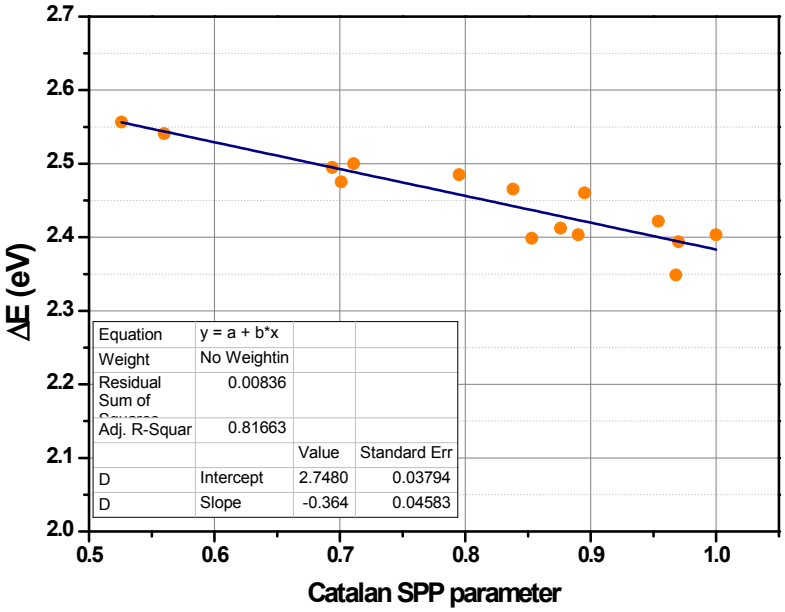
$y = -0.222\ x + 2.8231$ $R^2 = 0.571$

Compound I



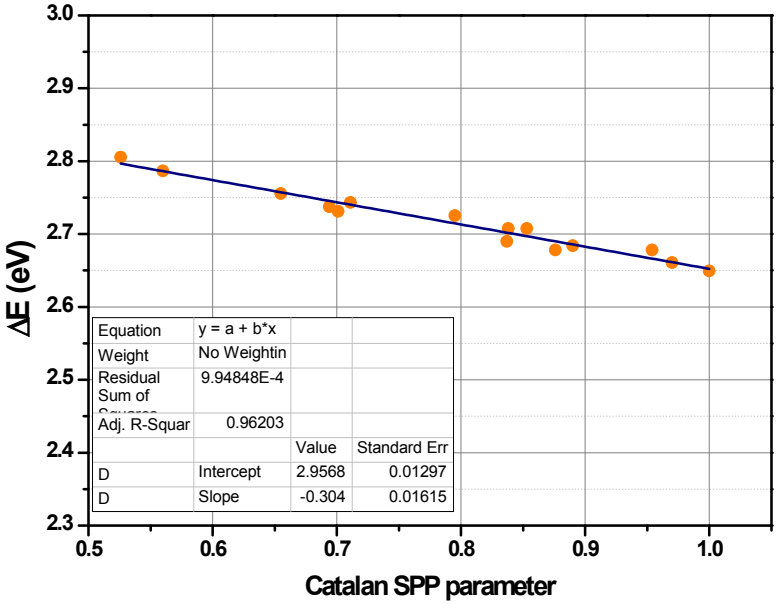
$y = -0.297\ x + 2.9246$ $R^2 = 0.896$

Compound J



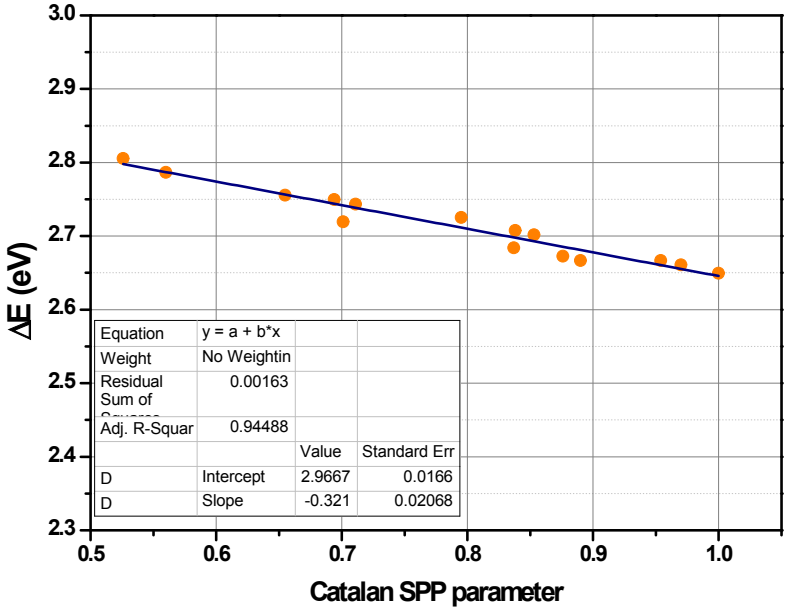
$y = -0\ 364\ x + 2.7480$ $R^2 = 0.817$

Compound K



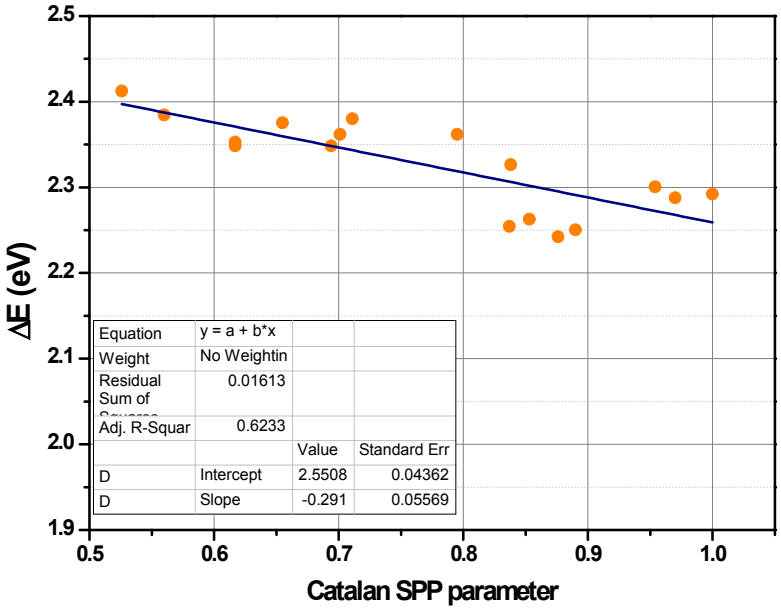
$y = -0.304\ x + 2.9568$ $R^2 = 0.962$

Compound L



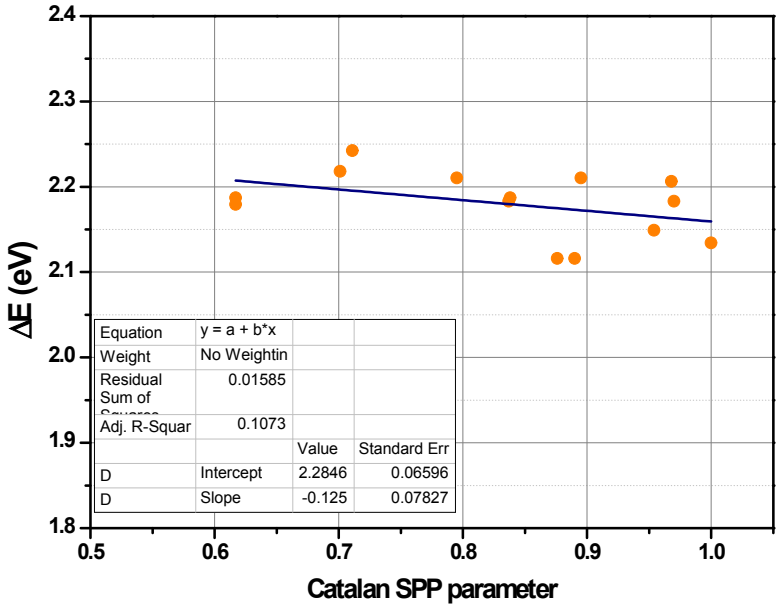
$y = -0.321\ x + 2.967$ $R^2 = 0.944$

Compound M



$y = -0.291\ x + 2.5508$ $R^2 = 0.623$

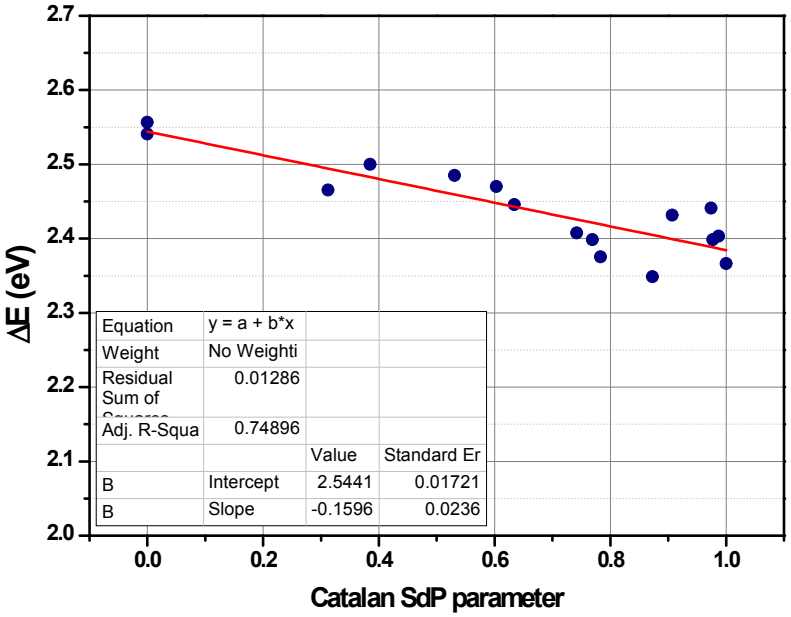
Compound N



$y = -0.125\ x + 2.2846$ $R^2 = 0.107$

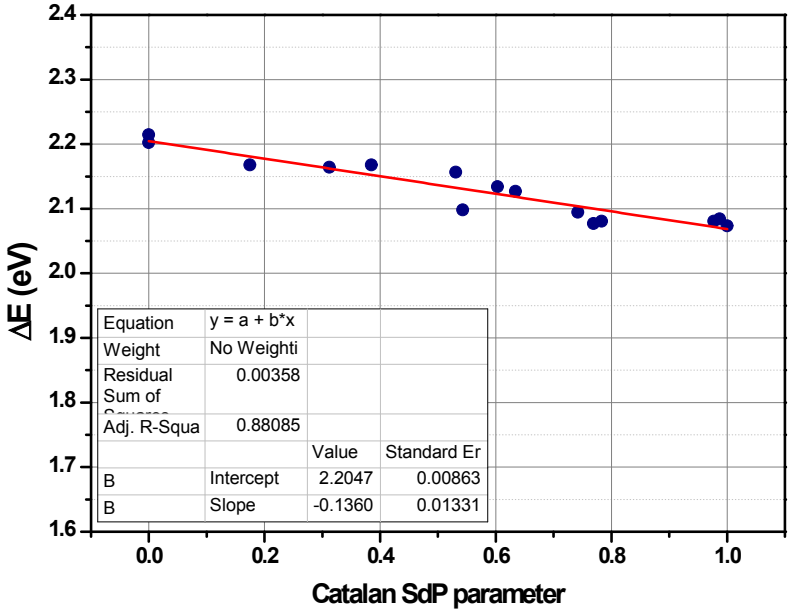
Position of the absorption maxima of A-N in 23 solvents of different polarities vs. the Catalan solvent dipolarity (SdP) scale

Compound A



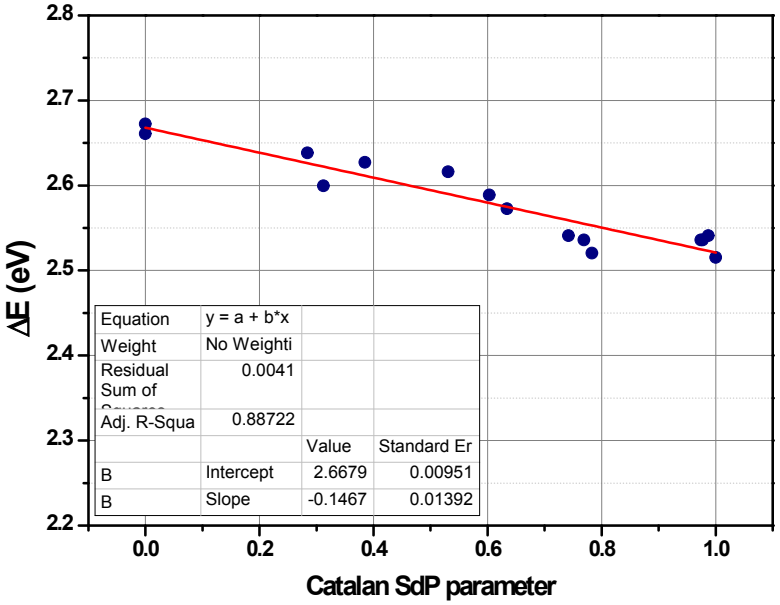
$y = -0.1596\ x + 2.5441$ $R^2 = 0.749$

Compound B



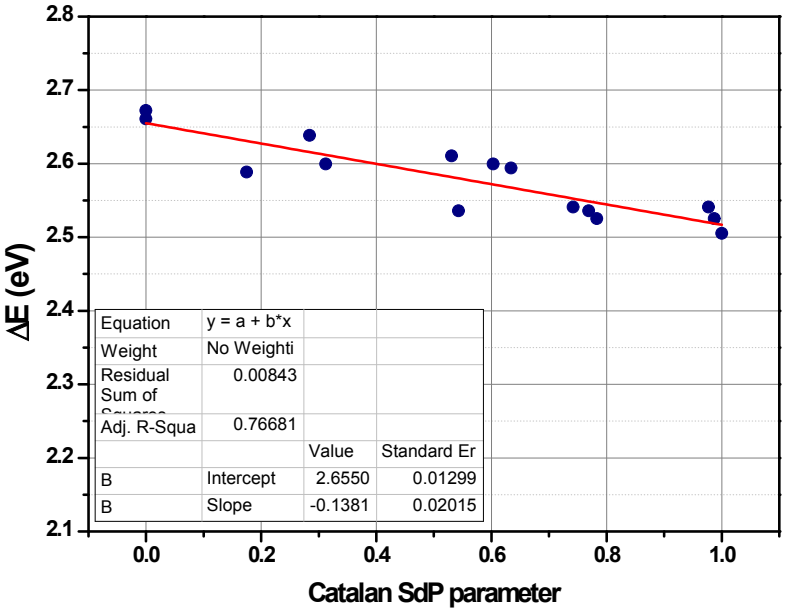
$y = -0.1360\ x + 2.2047$ $R^2 = 0.880$

Compound C



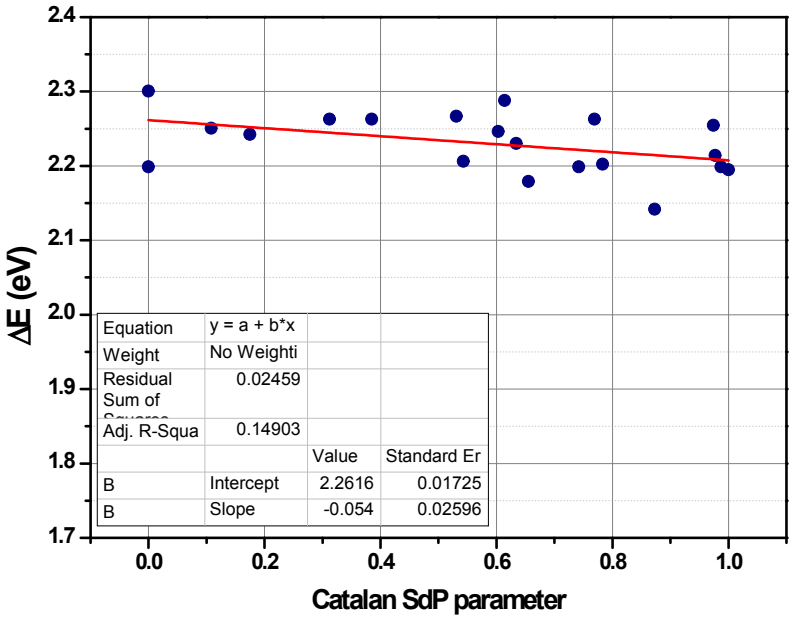
$y = -0.1467\ x + 2.6679$ $R^2 = 0.887$

Compound D



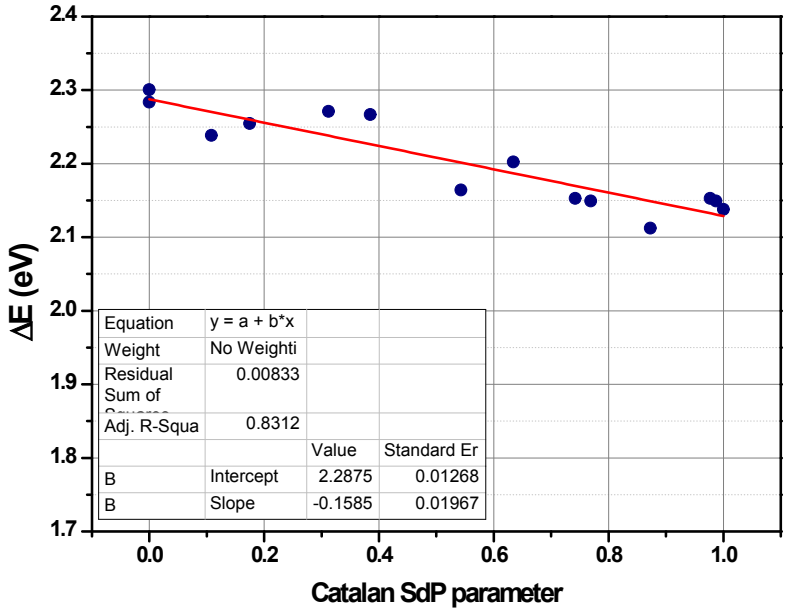
$y = -0.1381\ x + 2.6550$ $R^2 = 0.766$

Compound E



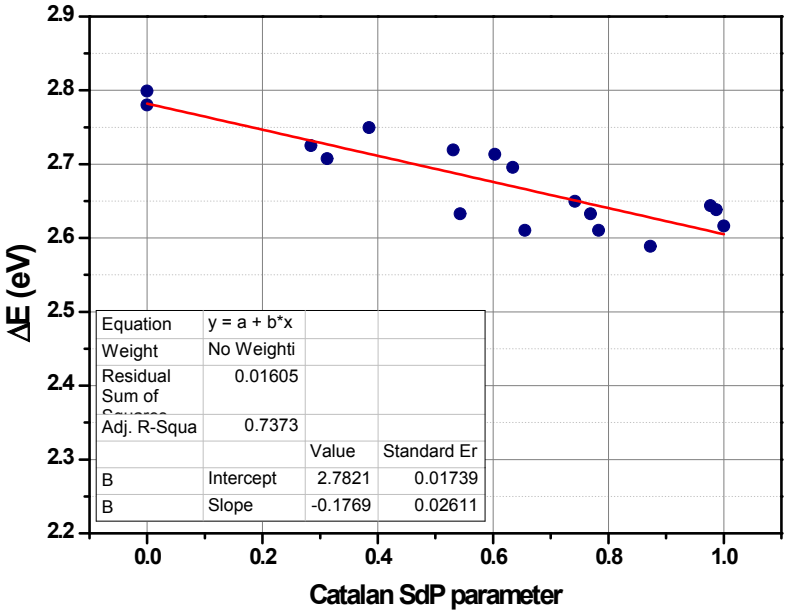
$y = -0.054\ x + 2.2616$ $R^2 = 0.149$

Compound F



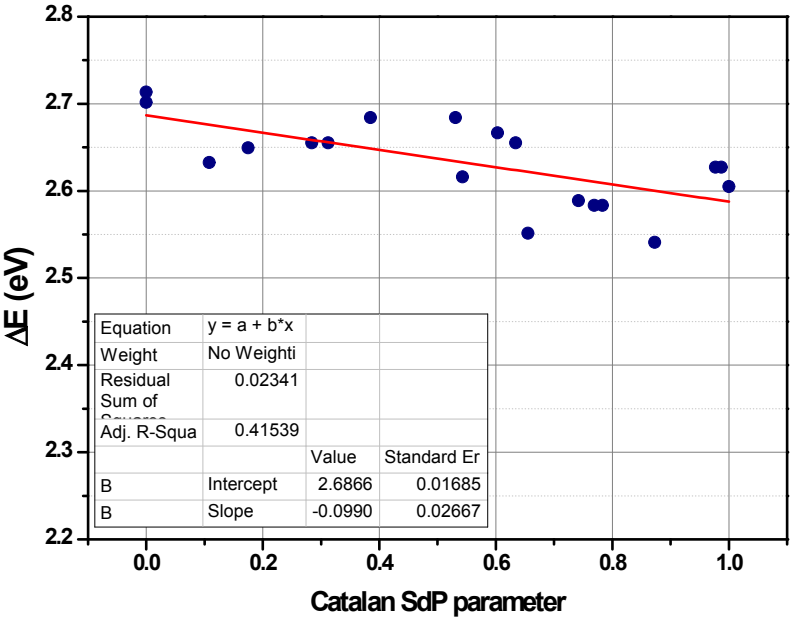
$y = -0.1585\ x + 2.2875$ $R^2 = 0.831$

Compound G



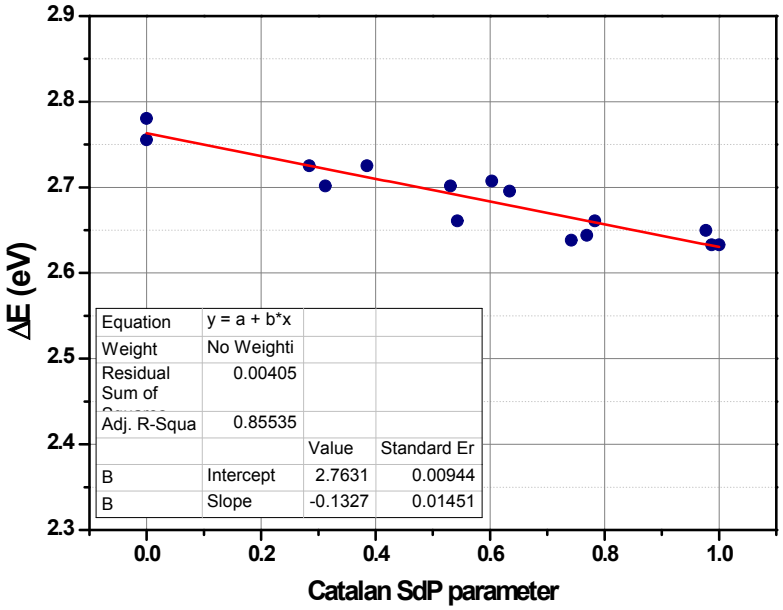
$y = -0.1769\ x + 2.7821$ $R^2 = 0.737$

Compound H



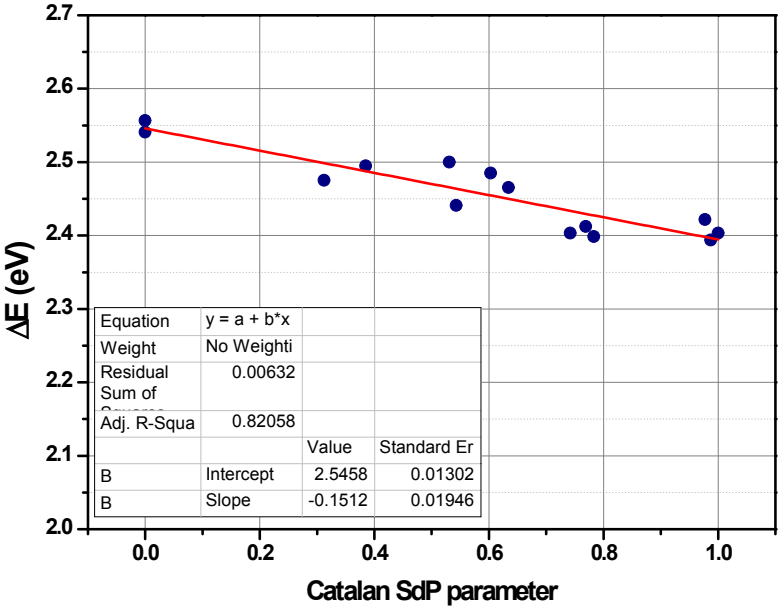
$y = -0.099\ x + 2.6866$ $R^2 = 0.415$

Compound I



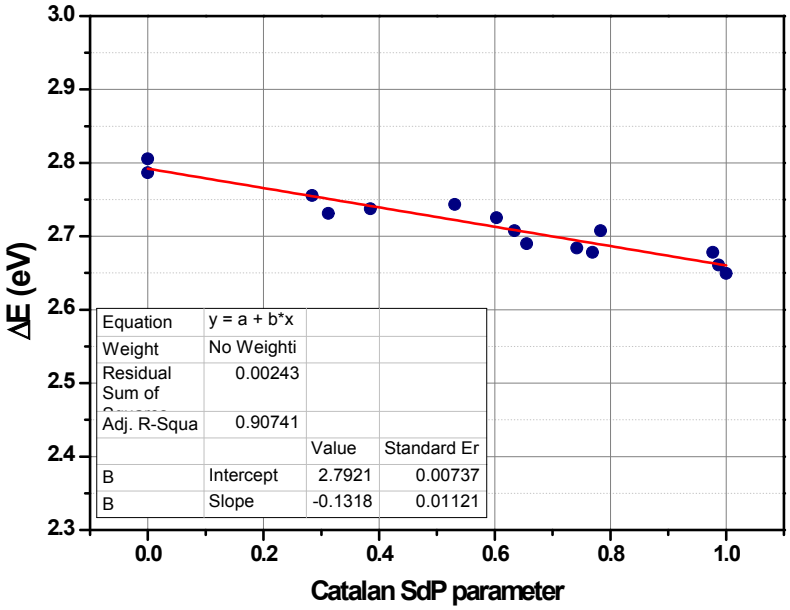
$y = -0.1327\ x + 2.7631$ $R^2 = 0.855$

Compound J



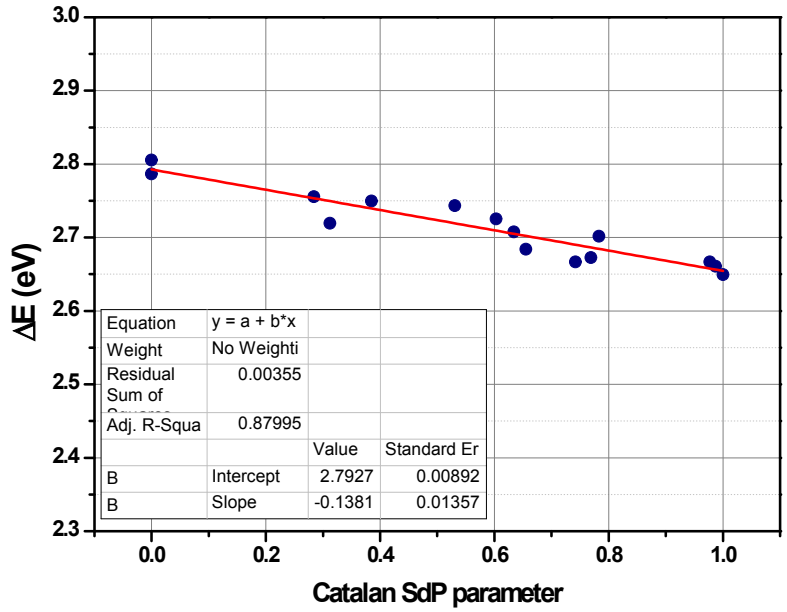
$y = -0\ 1512\ x + 2.5458$ $R^2 = 0.820$

Compound K



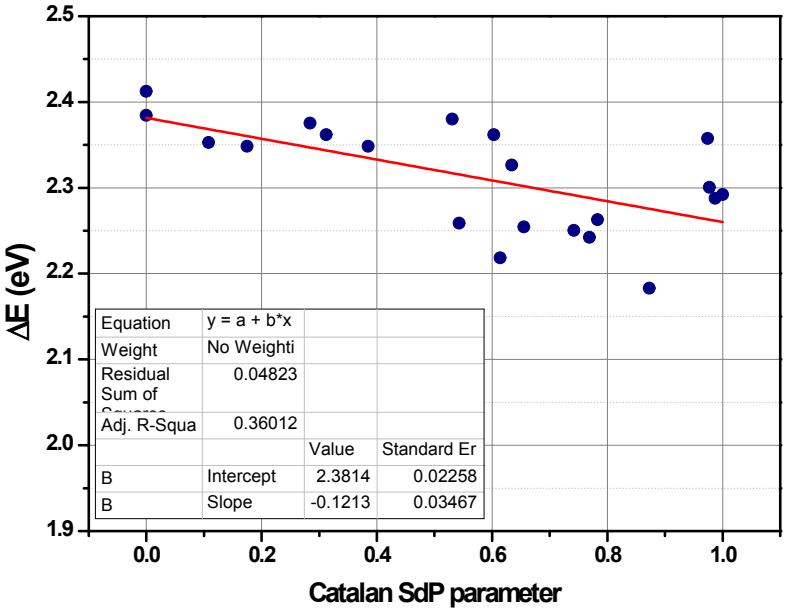
$y = -0.1318\ x + 2.7921$ $R^2 = 0.907$

Compound L



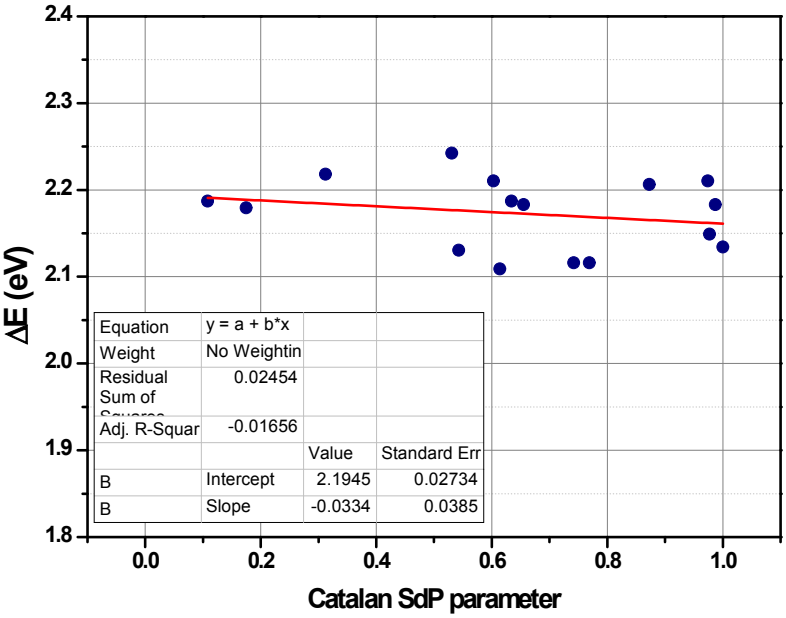
$y = -0.1381\ x + 2.7927$ $R^2 = 0.880$

Compound M



$y = -0.1213\ x + 2.3814$ $R^2 = 0.360$

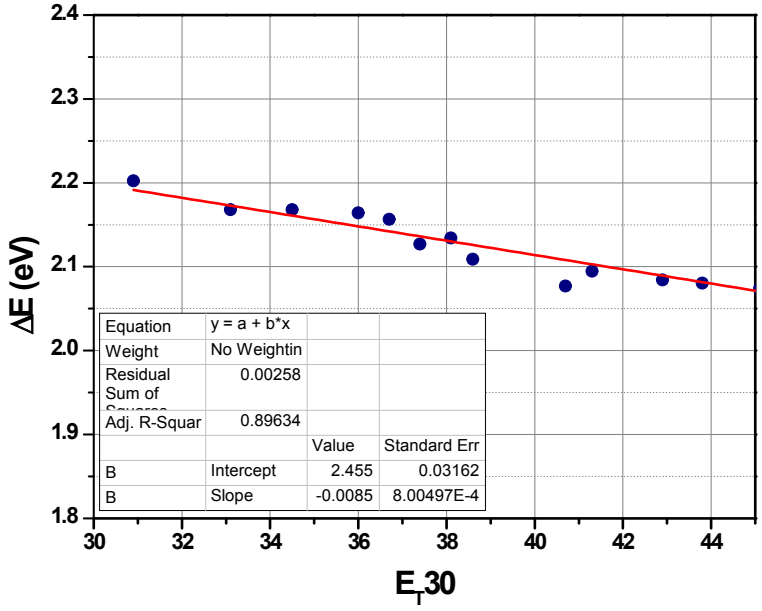
Compound N



$y = -0.0334\ x + 2.1945$ $R^2 = 0.016$

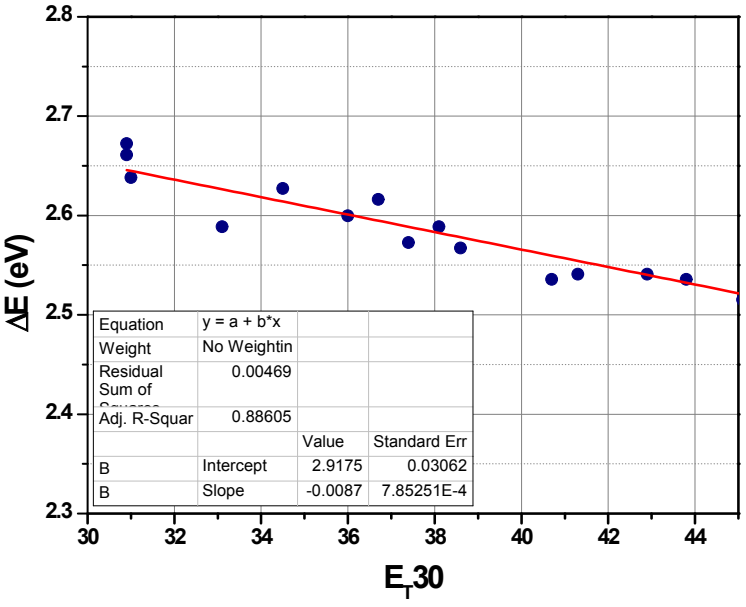
Position of the absorption maxima of dyes B-D, G-J and M in 23 solvents of different polarities vs. E_T30

Compound B



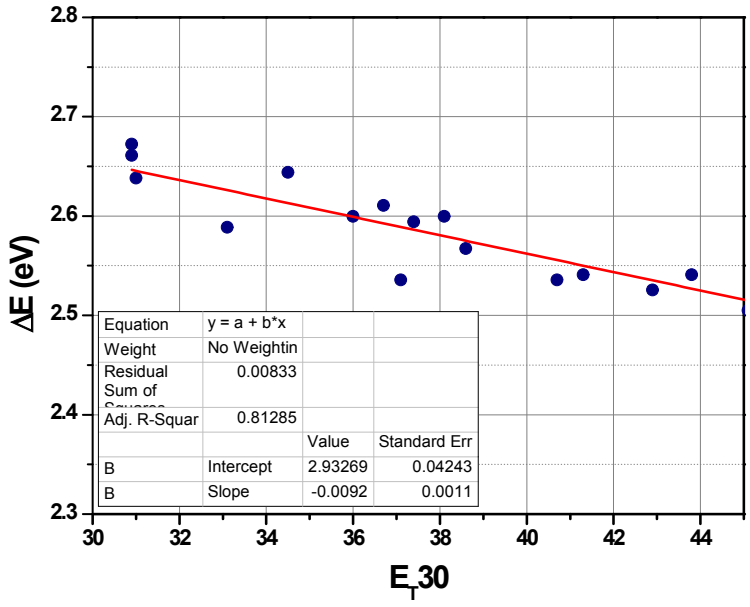
$y = -0.0085\ x + 2.455$ $R^2 = 0.896$

Compound C



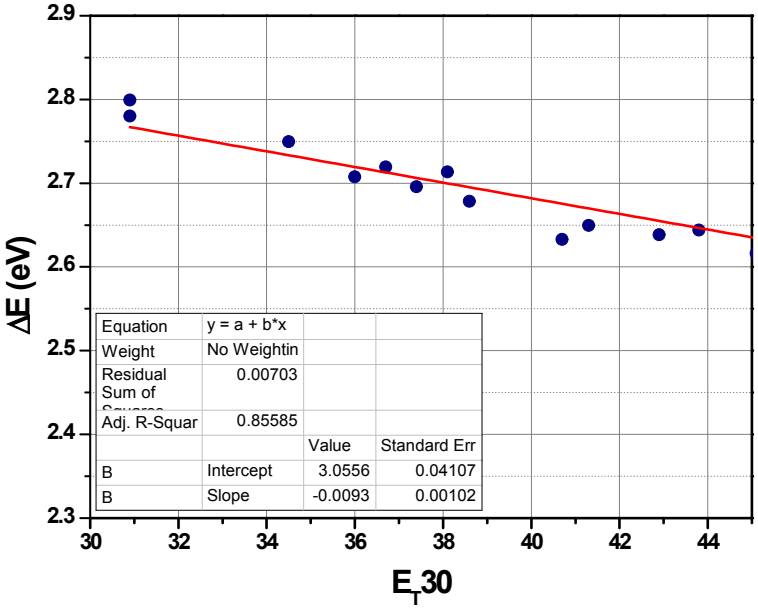
$y = -0.0087\ x + 2.917$ $R^2 = 0.886$

Compound D



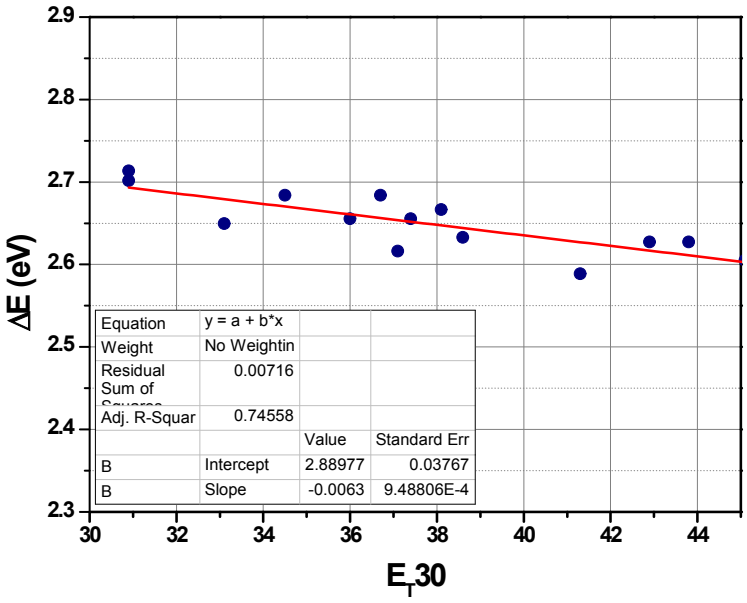
$y = -0.0092\ x + 2.93269$ $R^2 = 0.813$

Compound G



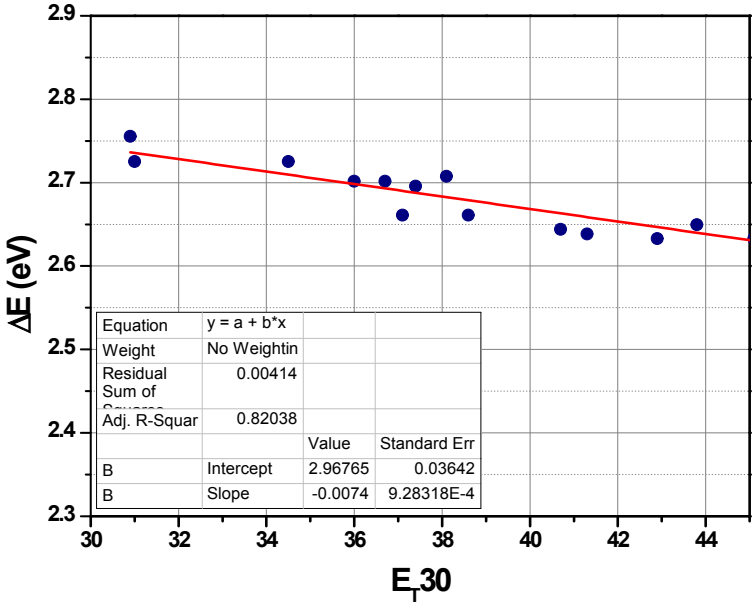
$y = -0.0093\ x + 3.0556$ $R^2 = 0.855$

Compound H



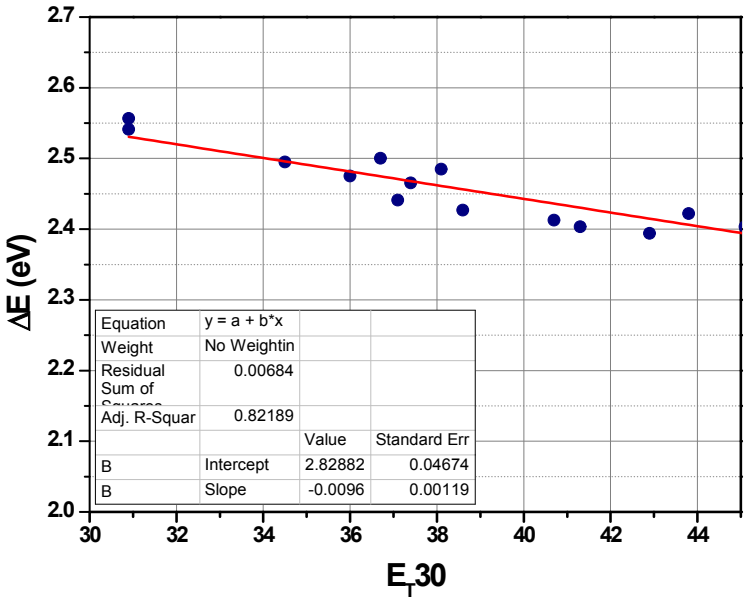
$y = -0.0063\ x + 2.88977$ $R^2 = 0.745$

Compound I



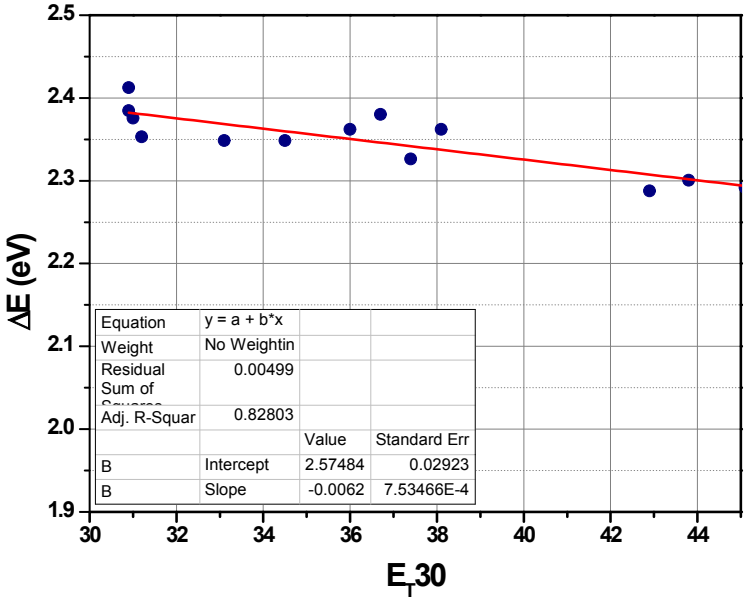
$y = -0.0074\ x + 2.96765$ $R^2 = 0.820$

Compound J



$y = -0.0096\ x + 2.82882$ $R^2 = 0.821$

Compound M



$y = -0.0062\ x + 2.57484$ $R^2 = 0.828$

Table S1. Summary of the optical properties of compounds **A-N** in twenty-three solvents, values of the Kamlet and Taft parameters π* and values of the Catalan solvent polarizability (SP), solvent dipolarity (SdP) and solvent polarity/polarizability (SPP) parameters

compounds	**π*[1]**	**SP**	**SdP**	**SPP**	**A**	**B**	**C**	**D**	**E**	**F**	**G**	**H**
acetone	0.71	0.651	0.907	0.881	510	585	456	456	553	546	456	466
acetonitrile	0.75	0.645	0.974	0.895	508	579	489	476	550	545	461	466
AcOEt	0.54	0.656	0.603	0.795	502	581	479	477	552	551	457	465
anisole	0.73	0.82	0.543	0.854	516	591	491	489	562	573	471	474
butanol	0.47	0.674	0.655	0.837	525	604	498	497	569	nd[3]	475	486
chloroform	0.78	0.783	0.614	0.786	526	610	495	495	542	583	478	489
cyclohexane	0.00	0.683	0	0.557	488	563	466	466	564	543	446	459
1,2-dichloroethane	0.81	0.771	0.742	0.890	515	592	488	488	564	576	468	479
dichloromethane	0.82	0.761	0.769	0.876	517	597	489	489	548	577	471	480
diethyl carbonate	0.45	0.653	0.531	0.711	499	575	474	475	547	546	456	462
diethyl ether	0.27	0.617	0.385	0.694	496	572	472	469	548	547	451	462
diglyme	0.64			0.777	508	588	483	483	559	574	463	471
1,4-dioxane	0.55	0.737	0.312	0.701	503	573	477	477	548	546	458	467
dimethylacetamide	0.88	0.763	0.987	0.970	516	595	488	491	564	577	470	472
dimethylformamide	0.87	0.759	0.977	0.954	517	596	489	488	560	576	469	472
DMSO	1.00	0.83	1	1.000	524	598	493	495	565	580	474	476
ethanol	0.54	0.633	0.783	0.853	522	596	492	491	563	nd[3]	475	480
heptane	-0.08	0.635	0	0.526	485	560	464	464	539	539	443	457
nitrobenzene	1.01	0.891	0.873	0.968	528	617	500	502	579	587	479	488
THF	0.58	0.714	0.634	0.838	507	583	482	478	556	563	460	467
toluene	0.54	0.66	0.108	0.617	509	581	483	483	551	554	464	471
triethylamine	0.14	0.782	0.284	0.655	-	nd[3]	470	470	nd[3]	nd[3]	455	467
p-xylene	0.43	0.778	0.175	0.617	508	572	479	479	553	550	462	468

[1] Kamlet and Taft parameters [2] Position of the ICT bands are given in nm [3] nd : not determined

Table S2. Summary of the optical properties of compounds **A-N** in twenty-three solvents, values of the Kamlet and Taft parameters π* and values of the Catalan solvent polarizability (SP), solvent dipolarity (SdP) and solvent polarity/polarizability (SPP) parameters

compounds	π*[1]	SP	SdP	SPP	I	J	K	L	M	N
acetone	0.71	0.651	0.907	0.881	462	460	456	457	534	562
acetonitrile	0.75	0.645	0.974	0.895	463	504	455	456	526	561
AcOEt	0.54	0.656	0.603	0.795	458	499	455	455	525	561
anisole	0.73	0.82	0.543	0.854	466	508	464	464	549	582
butanol	0.47	0.674	0.655	0.837	474	521	461	462	550	568
chloroform	0.78	0.783	0.614	0.786	475	524	466	468	559	588
cyclohexane	0.00	0.683	0	0.557	450	488	445	445	520	nd[3]
1,2-dichloroethane	0.81	0.771	0.742	0.890	470	516	462	465	551	586
dichloromethane	0.82	0.761	0.769	0.876	469	514	463	464	553	586
diethyl carbonate	0.45	0.653	0.531	0.711	459	496	452	452	521	553
diethyl ether	0.27	0.617	0.385	0.694	455	497	453	451	528	nd[3]
diglyme	0.64			0.777	466	511	461	461	545	569
1,4-dioxane	0.55	0.737	0.312	0.701	459	501	454	456	525	559
dimethylacetamide	0.88	0.763	0.987	0.970	471	518	466	466	542	568
dimethylformamide	0.87	0.759	0.977	0.954	468	512	463	465	539	577
DMSO	1.00	0.83	1	1.000	471	516	468	468	541	581
ethanol	0.54	0.633	0.783	0.853	466	517	458	459	548	nd[3]
heptane	-0.08	0.635	0	0.526	446	485	442	442	514	nd[3]
nitrobenzene	1.01	0.891	0.873	0.968	481	528	473	473	568	562
THF	0.58	0.714	0.634	0.838	460	503	458	458	533	567
toluene	0.54	0.66	0.108	0.617	465	506	459	460	527	567
triethylamine	0.14	0.782	0.284	0.655	455	nd[3]	450	450	522	nd[3]
p-xylene	0.43	0.778	0.175	0.617	464	504	458	459	528	569

[1] Kamlet and Taft parameters [2] Position of the ICT bands are given in nm. [3] nd : not determined

Results of the linear correlation analyses

The position of the UV/Vis absorption maxima with regard to the dipolarity/polarizability π* can be interpreted using a simplified version of the Kamlet–Taft equation :

$$\nu_{max}\,(cm^{-1}) = \nu_{max,0}\,(cm^{-1}) + s\pi^*$$

Table S3. Solvent-independent correlation coefficient *s* of the Kamlet-Taft parameters π*

Compounds	$\nu_{max,0}$	*s*	R^2
A	2.5512	-0.1774	0.951
B	2.20814	-0.14194	0.961
C	2.6676	-0.1640	0.908
D	2.67496	-0.1729	0.903
E	2.2912	-0.1091	0.733
F	2.29218	-0.1669	0.864
G	2.7914	-0.1762	0.898
H	2.71551	-0.1234	0.731
I	2.76451	-0.1503	0.838
J	2.54916	-0.1606	0.871
K	2.7872	-0.1434	0.890
L	2.78813	-0.1506	0.902
M	2.40304	-0.1597	0.594
N	2.24081	-0.1071	0.286

The position of the UV/Vis absorption maxima with regard to the dipolarity/polarizability π* can also be interpreted using a Catalan parameters, namely, the solvent dipolarity (SdP) and the solvent polarity/polarizability (SPP) using the following equations :

$$\nu_{max}\,(cm^{-1}) = \nu_{max,01}\,(cm^{-1}) + a \times SdP$$

$$\nu_{max}\,(cm^{-1}) = \nu_{max,02}\,(cm^{-1}) + b \times SPP$$

Table S4. Solvent-independent correlation coefficients a and b of the Catalan parameters SdP and SPP.

Compounds	$\nu_{max,01}$	*a*	R^2	$\nu_{max,02}$	*b*	R^2
A	2.5441	-0.1596	0.749	2.7599	-0.386	0.908
B	2.2047	-0.1360	0.880	2.3646	-0.292	0.977
C	2.6679	-0.1467	0.887	2.8506	-0.337	0.955
D	2.6550	-0.1381	0.766	2.8429	-0.340	0.723
E	2.2616	-0.054	0.149	2.3888	-0.195	0.775
F	2.2875	-0.1585	0.831	2.5013	-0.369	0.843
G	2.7821	-0.1769	0.737	2.9907	-0.374	0.877
H	2.6866	-0.099	0.415	2.8231	-0.222	0.571
I	2.7631	-0.1327	0.855	2.9246	-0.297	0.896
J	2.5458	-0 1512	0.820	2.7480	-0 364	0.817
K	2.7921	-0.1318	0.907	2.9568	-0.304	0.962
L	2.7927	-0.1381	0.880	2.967	-0.321	0.944
M	2.3814	-0.1213	0.360	2.5508	-0.291	0.623
N	2.1945	-0.0334	0.016	2.2846	-0.125	0.107

The position of the UV/Vis absorption maxima with regard to the E_T30 parameters can be interpreted using a simplified version of the Reichardt equation :

$$\nu_{max}\,(cm^{-1}) = \nu_{max,0}\,(cm^{-1}) + c \times E_T30$$

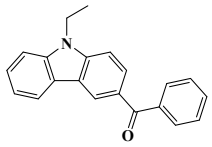
Table S3. Solvent-independent correlation coefficient c of the Reichardt parameters E_T30.

Compounds	$\nu_{max,01}$	*c*	R^2
B	2.455	-0.0085	0.896
C	2.917	-0.0087	0.886
D	2.93269	-0.0092	0.813
G	3.0556	-0.0093	0.855
H	2.88977	-0.0063	0.745
I	2.96765	-0.0074	0.820
J	2.82882	-0.0096	0.821
M	2.57484	-0.0062	0.828

Experimental part

All reagents and solvents were purchased from Aldrich, Alfa Aesar or TCI Europe and used as received without further purification. Mass spectroscopy was performed by the Spectropole of Aix-Marseille University. ESI mass spectral analyses were recorded with a 3200 QTRAP (Applied Biosystems SCIEX) mass spectrometer. The HRMS mass spectral analysis was performed with a QStar Elite (Applied Biosystems SCIEX) mass spectrometer. Elemental analyses were recorded with a Thermo Finnigan EA 1112 elemental analysis apparatus driven by the Eager 300 software. ^{1}H and ^{13}C NMR spectra were determined at room temperature in 5 mm o.d. tubes on a Bruker Avance 400 spectrometer of the Spectropole: ^{1}H (400 MHz) and ^{13}C (100 MHz). The ^{1}H chemical shifts were referenced to the solvent peak $CDCl_3$ (7.26 ppm) and the ^{13}C chemical shifts were referenced to the solvent peak $CDCl_3$ (77 ppm). Fourier-transformation infrared(FT-IR) spectra were recorded using a PerkinElmer Spectrum Two FT-IR Spectrometer with an ATR accessory. Position of peaks are given in cm^{-1}.

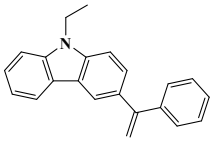
Synthesis of (9-ethyl-9*H*-carbazol-3-yl)(phenyl)methanone



Chemical Formula: $C_{21}H_{17}NO$
Molecular Weight: 299.3730

$AlCl_3$ (3.60 g, 27 mmol, M = 133.34 g/mol) was added by portion to a cooled solution of 9-ethyl-9*H*-carbazole (5.27 g, 27 mmol, M = 195.26 g/mol) and benzoyl chloride (3.80 g, 3.13 mL, 27 mmol, M = 140.57 g/mol, d = 1.211) dissolved in dry dichloromethane (DCM) (25 mL, stabilized with amylene). The solution was stirred at room temperature overnight. The solution was quenched onto ice. The solution was extracted several times with DCM. The organic phases were combined, dried over magnesium sulfate and the solvent removed under reduced pressure. The residue was purified by column chromatography (SiO2) using a mixture of DCM/pentane 1/1 as the eluent (6.79 g, 84% yield). ^{1}H NMR ($CDCl_3$) δ : 1.49 (t, 3H, J = 7.2 Hz), 4.42 (q, 2H, J = 7.2 Hz), 7.29 (td, 1H, J = 7.9 Hz, J = 1.0 Hz), 7.45-7.47 (m, 2H), 7.50-7.54 (m, 3H), 7.58-7.62 (m, 1H), 7.84-7.87 (m, 2H), 8.04 (dd, 1H, J = 8.6 Hz, J = 1.7 Hz), 8.12 (d, 1H, J = 7.7 Hz), 8.62 (d, 1H, J = 1.3 Hz); ^{13}C NMR ($CDCl_3$) δ : 13.8, 37.9, 108.0, 109.0, 120.0, 120.8, 122.6, 123.2, 124.1, 126.5, 128.2, 128.5, 128.6, 129.9, 131.6, 139.1, 140.7, 142.6, 196.6; HRMS (ESI MS) m/z: theor: 299.1310 found: 299.1313 ($[M]^{+\cdot}$ detected)

Synthesis of 9-ethyl-3-(1-phenylvinyl)-9*H*-carbazole

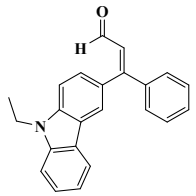


Chemical Formula: $C_{22}H_{19}N$
Molecular Weight: 297.4010

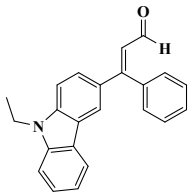
Magnesium (98%, 4.2 g, 170 mmol) and diethyl ether (20 mL) were stirred under nitrogen as iodomethane (99%, 25.0 g, 11 mL, 170 mmol, d = 2.28, M = 141.94 g/mol) was added dropwise over 20 min at such a rate as to maintain a continuous exotherm and gentle refluxing. The mixture was gently refluxed for 25 min by which time almost all of the magnesium had dissolved. The reaction mixture was protected from light and a solution of (9-ethyl-9*H*-carbazol-3-yl)(phenyl)methanone (9.89 g, 33 mmol, 1 eq., M = 299.37 g/mol) in toluene (280 mL) at room temperature was run in. The mixture was stirred

under nitrogen in the absence of light at ambient temperature for 22 h, before cautiously quenching with water (200 mL) to give a light blue emulsion, which was stirred for 15 min. A solution of ammonium chloride (30 g) and acetic acid (99%, 15 mL) in water (150 mL) was added and the whole stirred for 3.5 h. The aqueous phase was washed twice with toluene (50 mL) and the extracts combined with the organic phase, from which the solvent was removed under reduced pressure. It was used without any further purification in the next step (9.62 g, 98% yield). ^{1}H NMR ($CDCl_3$) δ : 1.32 (t, 3H, J = 7.2 Hz), 4.23 (q, 2H, J = 7.2 Hz), 5.39 (d, 1H, J = 1.3 Hz), 5.45 (d, 1H, J = 1.3 Hz), 7.07-7.17 (m, 2H), 7.24-7.31 (m, 4H), 7.34-7.40 (m, 4H), 7.96 (d, 1H, J = 7.7 Hz), 8.02 (d, 1H, J = 1.7 Hz); ^{13}C NMR ($CDCl_3$) δ : 13.8, 37.5, 107.9, 108.5, 113.0, 118.9, 120.2, 120.5, 122.8, 123.0, 125.3, 125.7, 126.3, 127.6, 128.1, 128.2, 128.4, 129.0, 132.5, 139.6, 140.3, 142.4, 150.7; HRMS (ESI MS) m/z: theor: 297.1517 found: 297.1518 ($[M]^{+\cdot}$ detected)

Synthesis of (Z/E)- 3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylacrylaldehyde



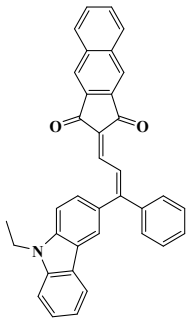
Chemical Formula: $C_{23}H_{19}NO$
Molecular Weight: 325.4110



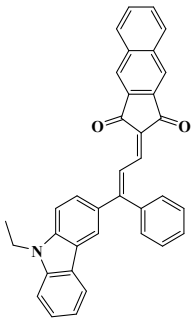
Chemical Formula: $C_{23}H_{19}NO$
Molecular Weight: 325.4110

To 2.5 mL (32.28 mmol) of dry DMF at 0°C was added 0.17 mL (0.288 g, 1.82 mmol) of $POCl_3$. After 15 min of stirring at 0°C, 9-ethyl-3-(1-phenylvinyl)-9*H*-carbazole (0.56 g, 1.88 mmol, M = 297.40 g/mol) in 2.5 mL dry DMF was added and the reaction mixture allowed to warm overnight to room temperature. The mixture was poured on water. Aq. 1M NaOH was added until formation of a precipitate. The resulting solid was filtered, dissolved in CH_2Cl_2, dried over $MgSO_4$, and the solvent evaporated under reduced pressure. Recrystallization from a mixture of pentane/CH_2Cl_2 afforded a light-yellow solid that was isolated under the form of a s-cis and s-trans mixture in a 2:1 ratio (0.54 g, 88% yield). Interestingly, by successive precipitations from a mixture of pentane/CH_2Cl_2, one of the isomers could be isolated in pure form. ^{1}H NMR ($CDCl_3$) δ : 1.43-1.52 (m, 9H), 4.34-4.46 (m, 6H), 6.62 (d, 1H, J = 7.9 Hz), 6.75 (d, 2H, J = 8.1 Hz), 7.22-7.29 (m, 3H), 7.37-7.55 (m, 28H), 8.01-8.09 (m, 6H), 9.52 (d, 2H, J = 8.1 Hz), 9.63 (d, 1H, J = 7.9 Hz); ^{13}C NMR ($CDCl_3$) δ : 13.8, 13.9, 37.8, 108.2, 108.5, 108.86, 108.90, 119.6, 119.7, 120.6, 120.7, 121.6, 122.6, 122.8, 122.9, 123.2, 123.7, 125.7, 126.39, 126.42, 126.6, 127.0, 127.3, 128.3, 128.5, 128.9, 129.2, 129.3, 130.3, 137.6, 140.50, 140.52, 140.7, 141.0, 141.4163.7, 163.8, 193.6, 194.1; HRMS (ESI MS) m/z: theor: 325.1467 found: 325.1465 ($[M]^{+\cdot}$ detected)

Synthesis of (Z/E)-2-(3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1*H*-cyclopenta[*b*]naphthalene -1,3(2*H*)-dione (A)



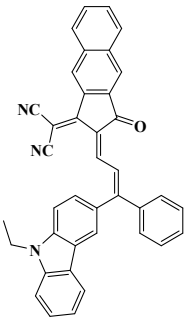
Chemical Formula: $C_{36}H_{25}NO_2$
Molecular Weight: 503.6010



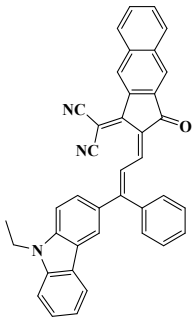
Chemical Formula: $C_{36}H_{25}NO_2$
Molecular Weight: 503.6010

3,3-*Bis*(4-(dimethylamino)phenyl)acrylaldehyde (1 g, 3.07 mmol, M = 325.41 g/mol) and 1*H*-cyclopenta[*b*]naphthalene-1,3(2*H*)-dione (0.61 g, 3.07 mmol, M = 196.20 g/mol) were suspended in ethanol (30 mL) and the flask was introduced into a preheated bath at 100°C. A few drops of piperidine were added and the solution was refluxed for 20 min. During reflux, a red precipitate formed. It was filtered off, washed several times with ethanol and dried under vacuum (0.88 g, 88% yield). ^{1}H NMR ($CDCl_3$) δ : 1.46-1.50 (m, 3.9H), 4.37-4.47 (m, 2.8H), 7.22-7.27 (m, 3H), 7.39-7.55 (m, 11.4H), 7.65-7.73 (m, 4.9H), 7.70-7.77 (m, 5.2H), 7.83 (d, 0.45H, J = 12.4 Hz), 8.03-8.09 (m, 4.3H), 8.20 (s, 1H), 8.41-8.59 (m, 2.6H), 8.57 (d, 0.4H, J = 12.4 Hz), 8.79 (d, 1H, J = 12.4 Hz); ^{13}C NMR ($CDCl_3$) δ :13.8, 13.9, 37.8, 108.4, 108.6, 108.8, 108.9, 119.6, 119.8, 120.9, 121.5, 122.7, 122.8, 123.1, 123.13, 123.4, 123.47, 123.52, 123.6, 124.0, 126.3, 126.4, 127.8, 128.46, 128.50, 128.57, 128.84, 128.88, 129.6, 130.1, 130.4, 131.3, 131.7, 136.2, 136.3, 136.4, 137.5, 137.6, 140.5, 140.6, 141.0, 141.6, 142.1, 144.4, 144.9, 164.8, 164.9, 190.3, 190.4, 190.7, 191.0; HRMS (ESI MS) m/z: theor: 503.1885 found: 503.1888 ($[M]^{+\cdot}$ detected) FT-IR (ATR): 1708.53, 1671.08, 1618.41, 1555.42, 1527.58, 1494.33, 1470.54, 1440.8, 1410.97, 1363.49, 1328.32, 1284.33, 1237.94, 1220.7, 1170.11, 1161.29, 1119.22, 1024.89, 1004.82, 962.71, 933.03, 904.91, 890.08, 800.17, 787.41, 766.46, 745.06, 733.1, 722.73, 698.33, 669.54, 631.93, 575.72, 538.84

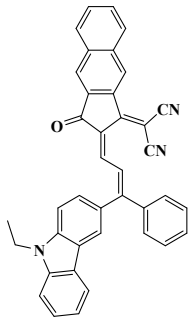
Synthesis of 2-((*Z*/*E*)-2-((*Z*/*E*)-3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-3-oxo-2,3-dihydro-1*H*-cyclopenta[*b*]naphthalen-1-ylidene)malononitrile (B)



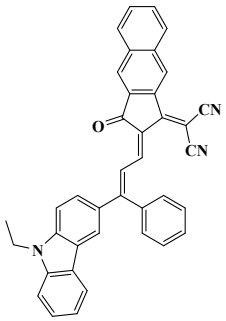
Chemical Formula: $C_{39}H_{25}N_3O$
Molecular Weight: 551.6490



Chemical Formula: $C_{39}H_{25}N_3O$
Molecular Weight: 551.6490



Chemical Formula: $C_{39}H_{25}N_3O$
Molecular Weight: 551.6490

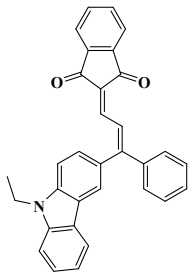


Chemical Formula: $C_{39}H_{25}N_3O$
Molecular Weight: 551.6490

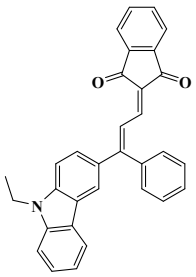
3,3-*Bis*(4-(dimethylamino)phenyl)acrylaldehyde (3 g, 9.21 mmol, M = 325.41 g/mol) (aldehyde 30:70 donc 3 eq.) and 2-(3-oxo-2,3-dihydro-1*H*-cyclopenta[*b*]naphthalen-1-ylidene)malononitrile (2.25 g, 9.21 mmol, M = 244.25 g/mol) were suspended in ethanol (30 mL) and the flask was introduced into a preheated bath at 100°C. A few drops of piperidine were added and the solution was refluxed for 20 min. During reflux, a purple precipitate formed. It was filtered off, washed several times with ethanol and dried under vacuum (4.67 g, 92% yield). ^{1}H NMR ($CDCl_3$) δ : 1.48 (t, 3H, J = 7.1 Hz), 1.53 (t, 1.4H, J

= 7.1 Hz), 4.37 (q, 2H, J = 7.1 Hz), 4.46 (q, 0.71H, J = 7.1 Hz), 7.24-7.29 (m, 2H), 7.36-7.61 (m, 12H), 7.66-7.76 (m, 4H), 7.90-8.11 m, 4.5H), 8.23 (s, 1H), 8.35-8.42 (m, 2.3H), 8.58 (d, 0.3H, J = 12.1 Hz), 8.88 (d, 0.3H, J = 12.1 Hz), 9.06-9.10 (m, 1.3H), 9.11 (d, 1H, J = 12.2 Hz); ^{13}C NMR ($CDCl_3$) δ : 13.9, 37.9, 68.2, 69.0, 108.8, 108.89, 108.92, 109.1, 114.2, 114.4, 115.2, 115.5, 119.6, 120.1, 120.9, 121.0, 122.3, 122.9, 123.0, 123.38, 123.42, 123.57, 123.60, 124.2, 124.3, 124.5, 126.4, 126.5, 126.6, 126.9, 128.2, 128.3, 128.6, 128.8, 129.4, 129.5, 129.6, 129.7, 129.8, 130.1, 130.2, 130.5, 130.7, 130.9, 131.0, 131.4, 133.5, 133.6, 134.6, 135.3, 135.4, 136.1, 136.2, 137.8, 140.56, 140.64, 141.5, 142.08, 142.14, 146.2, 146.9, 160.1, 160.3, 166.9, 189.2, 189.4; HRMS (ESI MS) m/z: theor: 551.1998 found: 551.2001 ([M]$^{+\cdot}$ detected) FT-IR (ATR) : 2213.49, 1685.97, 1624.20, 1549.26, 1538.02, 1512.75, 1469.68, 1441.35, 1410.69, 1364.83, 1337.21, 1285.07, 1221.29, 1170.58,1151.54, 1118.00, 999.88, 932.78, 889.52, 849.41, 799.66, 763.54, 744.93, 729.50, 701.74 ,671.99, 613.84 590.72, 547.93,

Synthesis of (*Z*/*E*)-2-(3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1*H*-indene-1,3(2*H*)-dione (C)



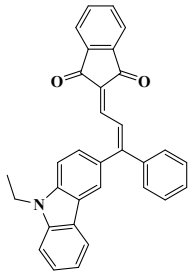
Chemical Formula: $C_{32}H_{23}NO_2$
Molecular Weight: 453.5410



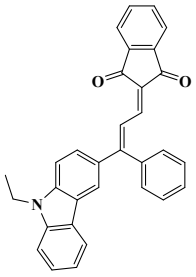
Chemical Formula: $C_{32}H_{23}NO_2$
Molecular Weight: 453.5410

3,3-*Bis*(4-(dimethylamino)phenyl)acrylaldehyde (1 g, 3.07 mmol, M = 325.41 g/mol) and indane-1,3-dione (0.450 g, 3.07 mmol, M = 146.14 g/mol) were suspended in ethanol (30 mL) and the flask was introduced into a preheated bath at 100°C. A few drops of piperidine were added and the solution was refluxed for 20 min. During reflux, a red precipitate formed. It was filtered off, washed several times with ethanol and dried under vacuum (1.14 g, 82% yeld). ^{1}H NMR ($CDCl_3$) δ : 1.39 (dt, J = 18.5, 7.2 Hz, 4H), 4.31 (dq, J = 21.6, 7.1 Hz, 2H), 7.15 (dd, J = 9.1, 5.6 Hz, 2H), 7.47 – 7.21 (m, 10H), 7.59 – 7.47 (m, 2H), 7.70 – 7.59 (m, 3H), 7.89 – 7.76 (m, 2H), 7.93 – 7.89 (m, 1H), 7.95 (t, J = 5.8 Hz, 1H), 8.07 (d, J = 1.5 Hz, 1H), 8.37 (d, J = 12.4 Hz, 1H), 8.57 (d, J = 12.6 Hz, 1H); ^{13}C NMR ($CDCl_3$) δ : 13.87, 13.95, 37.83, 108.55, 108.91, 119.77, 120.84, 121.03, 122.43, 122.61, 122.81, 123.05, 123.30, 126.34, 126.64, 127.59, 128.45, 129.39, 129.95, 131.18, 131.67, 134.58, 134.71, 138.32, 140.54, 140.78, 141.49, 142.15, 143.44, 163.80, 190.63, 191.22; HRMS (ESI MS) m/z: theor: 453.1729 found: 453.1731 ([M]$^{+\cdot}$ detected) FT-IR (ATR) : 1712.58, 1670.83, 1627.13, 1597.2, 1574.26, 1548.28, 1494.51; 1476.63, 1461.36, 1438.55, 1391.60, 1367.32, 1347.08, 1325.29, 1286.47, 1239.19 1221.45, 1190.61, 1148.45, 1128.22, 1088.93, 1060.50, 1022.79, 990.98, 956.22, 943.72, 931.88, 903.83, 882.97, 840.14, 810.28, 787.1, 772.1, 748.83, 725.84, 699.64, 674.03, 630.93, 579.83, 553.20, 535.43

Synthesis of (*Z*/*E*)-2-(3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1*H*-indene-1,3(2*H*)-dione (D)



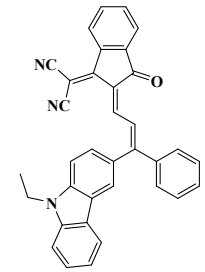
Chemical Formula: $C_{32}H_{23}NO_2$
Molecular Weight: 453.5410



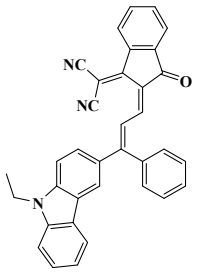
Chemical Formula: $C_{32}H_{23}NO_2$
Molecular Weight: 453.5410

For this synthesis of **D**, one of the isomers of 3,3-*bis*(4-(dimethylamino)phenyl)acrylaldehyde isolated in pure form was used. 3,3-*Bis*(4-(dimethylamino)phenyl)acrylaldehyde (1 g, 3.07 mmol, M = 325.41 g/mol) and indane-1,3-dione (0.450 g, 3.07 mmol, M = 146.14 g/mol) were suspended in ethanol (30 mL) and the flask was introduced into a preheated bath at 100°C. A few drops of piperidine were added and the solution was refluxed for 20 min. During reflux, a red precipitate formed. It was filtered off, washed several times with ethanol and dried under vacuum (1.24 g, 89% yield). ^{1}H NMR ($CDCl_3$) δ : 1.47 (t, 3H, J = 7.2 Hz), 4.40 (q, 2H, J = 7.2 Hz), 7.25 (td, 1H, J = 6.9 Hz, J = 1.2 Hz), 7.34-7.54 (m, 8H), 7.62 (d, 1H, J = 12.6 Hz), 7.66 (dd, 1H, J = 8.7 Hz, J = 1.8 Hz), 7.73-7.77 (m, 2H), 7.90-7.93 (m, 1H), 7.94-7.97 (m, 1H), 8.06 (d, 1H, J = 7.7 Hz), 8.17 (d, 1H, J = 1.6 Hz), 8.66 (d, 1H, J = 12.5 Hz); ^{13}C NMR ($CDCl_3$) δ : 13.9, 37.8, 108.5, 108.9, 119.8, 120.8, 121.0, 122.4, 122.6, 122.7, 122.8, 123.0, 123.3, 126.3, 126.4, 126.6, 127.6, 128.4, 129.4, 129.9, 131.2, 131.7, 134.6, 134.7, 138.3, 140.5, 140.8, 141.5, 142.1, 143.4, 163.8, 190.6, 191.2; HRMS (ESI MS) m/z: theor: 453.1729 found: 453.1731 ($[M]^{+\cdot}$ detected) FT-IR (ATR) : 1712.58, 1670.83, 1627.13, 1597.2, 1574.26, 1548.28, 1494.51; 1476.63, 1461.36, 1438.55, 1391.60, 1367.32, 1347.08, 1325.29, 1286.47, 1239.19 1221.45, 1190.61, 1148.45, 1128.22, 1088.93, 1060.50, 1022.79, 990.98, 956.22, 943.72, 931.88, 903.83, 882.97, 840.14, 810.28, 787.1, 772.1, 748.83, 725.84, 699.64, 674.03, 630.93, 579.83, 553.20, 535.43

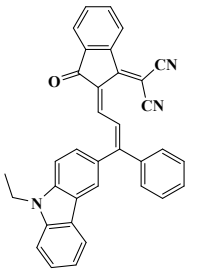
Synthesis of 2-((*Z*/*E*)-2-((*Z*/*E*)-3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-3-oxo-2,3-dihydro-1*H*-inden-1-ylidene)malononitrile (E)



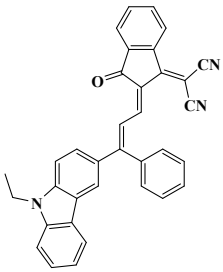
Chemical Formula: $C_{35}H_{23}N_3O$
Molecular Weight: 501.5890



Chemical Formula: $C_{35}H_{23}N_3O$
Molecular Weight: 501.5890



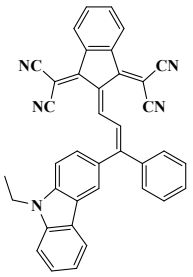
Chemical Formula: $C_{35}H_{23}N_3O$
Molecular Weight: 501.5890



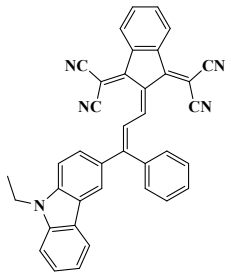
Chemical Formula: $C_{35}H_{23}N_3O$
Molecular Weight: 501.5890

2-(3-Oxo-2,3-dihydro-1*H*-inden-1-ylidene)malononitrile (0.330 g, 1.69 mmol, M = 194.19 g/mol) were suspended in ethanol (20 mL) and the flask was introduced into a preheated bath at 100°C. A few drops of DIPEA were added and the solution was refluxed. After cooling a violet precipitate formed. It was filtered off, washed several times with pentane and dried under vacuum (0.69 g, 82% yield). ^{1}H NMR ($CDCl_3$) δ : 1.45-1.52 (m, 4.21H), 4.36-4.45 (m, 2.80), 7.23-7.28 (m, 2H), 7.38-7.55 (m, 11.7H), 7.71-7.73 (m, 3.8H), 7.91-7.93 (m, 1.4H), 8.03-8.06 (m, 1.8H), 8.20 (s, 1H), 8.36 (d, 1H, J = 12.1 Hz), 8.52 (d, 0.4H, J = 12.0 Hz), 8.61-8.64 (m, 1.40H), 8.77 (d, 0.4H, J = 12.0 Hz), 9.05 (d, 1H, J = 12.1 Hz); ^{13}C NMR ($CDCl_3$) δ : 13.8, 13.9, 37.9, 69.5, 70.1, 108.7, 108.8, 108.9, 109.0, 113.8, 113.9, 114.7, 114.9, 119.6, 120.1, 120.9, 121.0, 121.8, 122.9, 123.1, 123.4, 123.5, 123.8, 125.1, 125.2, 126.4, 126.5, 126.6, 128.1, 128.2, 128.6, 128.8, 129.69, 129.71, 130.5, 130.7, 131.0, 131.4, 134.2, 123.3, 134.9, 135.0, 137.4, 137.5, 137.8, 139.9, 139.9, 140.6, 140.62, 141.4, 142.0, 142.1, 145.8, 146.3, 160.3, 160.4, 166.1, 166.2, 189.3, 189.6; HRMS (ESI MS) m/z: theor: 501.1841 found: 501.1838 ($[M]^{+\cdot}$ detected) FT-IR (ATR) 2221.5 0, 1700.50, 1592.79, 1563.68, 1549.89, 1533.92, 1491.69, 1470.52, 1438.75, 1386.58, 1369.47, 1359.02, 1344.11, 1250.24, 1231.16, 1202.31, 1183.11, 1153.05, 1133.62, 1122.99, 1091.34, 1025.56, 993.81, 943.58, 923.02, 890.78, 826.31, 795.62, 771.02, 750.46, 740.52, 724.81, 710.86, 671.46, 631.87, 604.8, 579.34, 557.48, 539.64

Synthesis of (*Z*/*E*)-2,2'-(2-(3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1*H*-indene-1,3(2*H*)-diylidene)dimalononitrile (F)



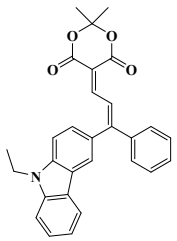
Chemical Formula: $C_{38}H_{23}N_5$
Molecular Weight: 549.6370



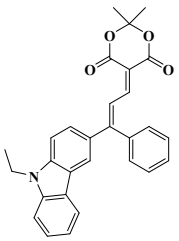
Chemical Formula: $C_{38}H_{23}N_5$
Molecular Weight: 549.6370

2,2'-(1*H*-indene-1,3(2*H*)-diylidene)dimalononitrile (1.22 g, 5.02 mmol, M = 242.24 g/mol), 3-and (9-ethyl-9*H*-carbazol-3-yl)-3-(4-methoxyphenyl)acrylaldehyde (1.78 g, 5.02 mmol, M = 355.44 g/mol) were dissolved in acetic anhydride (20 mL) and the solution was refluxed for two hours. After cooling, the solvent was removed under reduced pressure. The residue was suspended in a minimum of CH_2Cl_2 and addition of pentane precipitated a solid that was filtered off, washed several times with pentane and dried under vacuum (2.35 g, 85% yield). ^{1}H NMR ($CDCl_3$) δ : 1.44-.153 (5.51H), 4.35-4.47 (m, 3H), 7.23-7.37 (m, 2H), 7.36-7.50 (m, 12.4H), 7.68-7.75 (m, 4H), 7.90-7.94 (m, 1.6H), 8.00-8.07 (m, 3H), 8.20 (d, 1H, J = 1.7 Hz), 8.36 (d, 1H, J = 12.2 Hz), 8.52 (d, 0.5H, J = 12.2 Hz), 8.60-8.63 (m, 1.5H), 8.77 (d, 0.5H, J = 12.2 Hz), 9.04 (d, 1H, J = 12.2 Hz); ^{13}C NMR ($CDCl_3$) δ : 13.8, 13.9, 54.8, 69.4, 70.1, 104.5, 108.7, 109.0, 118=3.8, 113.9, 114.9, 118.4, 119.6, 119.8, 120.1, 120.9, 121.0, 121.8, 122.3, 122.9, 123.0, 123.2, 123.5, 123.6, 123.8, 124.2, 125.1, 126.4, 126.6, 128.1, 128.6, 128.8, 129.7, 129.9, 130.5, 130.7, 131.0, 131.4, 134.2, 134.3, 135.0, 137.5, 137.8, 138.5, 139.9, 140.6, 131.3, 142.0, 142.1, 145.8, 146.3, 160.0, 160.3, 166.1, 189.3, 189.6; HRMS (ESI MS) m/z: theor: 549.1953 found: 549.1952 ([M]$^{+\cdot}$ detected) FT-IR (ATR): 2221.1, 2189.27, 1699.7, 1592.41, 1563.21, 1549.53, 1533.11, 1489.40, 1455.56, 1438.24, 1386.20, 1368.93, 1358.69, 1341.93, 1249.141230.82, 1201.42, 1181.90, 1151.16, 1133.10, 1121.38, 1090.59, 1025.31, 993.30, 944.36, 936.10, 922.58, 890.52, 825.94, 804.78, 793.24, 770.24, 749.01 724.46, 710.41, 671.03, 631.65, 604.67, 578.90, 559.28, 538.94

Synthesis of (Z/E)-5-(3-(9-ethyl-9H-carbazol-3-yl)-3-phenylallylidene)-2,2-dimethyl-1,3-dioxane-4,6-dione (G)



Chemical Formula: $C_{29}H_{25}NO_4$
Molecular Weight: 451.5220

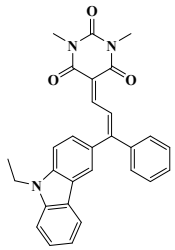


Chemical Formula: $C_{29}H_{25}NO_4$
Molecular Weight: 451.5220

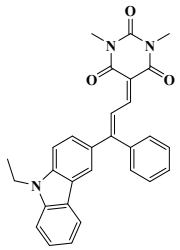
3-(9-Ethyl-9*H*-carbazol-3-yl)-3-(4-methoxyphenyl)acrylaldehyde (3.27 g, 9.21 mmol, M = 355.44 g/mol) and Meldrum acid (2,2-Dimethyl-1,3-dioxane-4,6-dione) (1.32 g, 9.21 mmol, M = 144.13 g/mol) were suspended in ethanol (30 mL) and a few drops of piperidine were added. The solution was refluxed overnight. The solution was concentrated under reduced pressure. The residue was dissolved in a minimum of ether and pentane was added. A precipitate formed. It was filtered off, washed several times with pentane and dried under vacuum (3.45 g, 83% yield). ^{1}H NMR ($CDCl_3$) δ : 1.45-1.54 (m, 5H), 1.77 (s, 9.3H), 4.36-4.46 (m, 3.3H), 7.24-7.28 (m, 1H), 7.32-7.44 (m, 6.6H), 7.46-7.56 (m, 7.0H), 7.65 (dd, 1H,

J = 8.7 Hz, J = 1.8 Hz), 7.96 (d, 0.5H, J = 1.8 Hz), 8.03-8.06 (m, 1.6H), 8.11 (d, 1H, J = 12.6 Hz), 8.14 (d, 1H, J = 1.4 Hz), 8.24 (s, 1.1H), 8.48 (d, 1H, J = 12.6 Hz); ^{13}C NMR ($CDCl_3$) δ : 13.8, 13.9, 27.64, 27.67, 37.9, 104.2, 104.3, 108.4, 108.7, 108.9, 109.0, 109.2, 110.5, 119.7, 120.0, 120.8, 120.9, 121.9, 122.7, 123.0, 123.1, 123.1, 123.3, 123.4, 124.2, 126.5, 126.5, 128.0, 128.3, 128.5, 128.6, 129.6, 130.4, 130.8, 131.2, 138.1, 140.5, 140.6, 141.3, 141.7, 141.9, 156.0, 156.7, 161.3, 161.5, 163.4, 163.5, 167.1, 167.2; HRMS (ESI MS) m/z: theor: 451.1784 found: 451.1786 ($[M]^{+\cdot}$ detected) FT-IR (ATR) : 1708.92, 1539.75, 1493.98, 1468.79, 1444.21, 1367.35, 1346.18, 1275.1, 1253.51, 1229.51, 1182.06, 1156.42, 1124.72, 1102.11, 1008.74, 92, 886.86, 808.95, 791.71, 768.43, 749.79, 735.71, 706.31, 694.11, 675.43, 633.75

Synthesis of (*Z*/*E*)-5-(3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1,3-dimethylpyrimidine-2,4,6(1*H*,3*H*,5*H*)-trione (H/I)



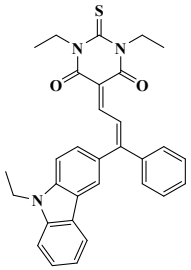
Chemical Formula: $C_{29}H_{25}N_3O_3$
Molecular Weight: 463.5370



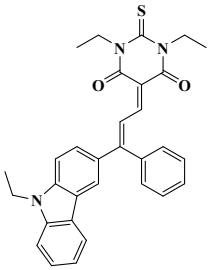
Chemical Formula: $C_{29}H_{25}N_3O_3$
Molecular Weight: 463.5370

1,3-Dimethylpyrimidine-2,4,6(1*H*,3*H*,5*H*)-trione (783 mg, 5.02 mmol, M = 156.14 g/mol) and 3,3-*bis*(4-(dimethylamino)phenyl)acrylaldehyde (1.63 g, 5.02 mmol, M = 325.41 g/mol) were dissolved in absolute ethanol (20 mL) and a few drops of piperidine were added. The solution was refluxed overnight. After cooling, the solvent was removed under reduced pressure. Addition of ether followed by pentane precipitated a blue solid that was filtered off. Interestingly, upon concentration of the solution, a second precipitate formed and was separated from the solution by filtration. Il was identified as being the isomer **I.** Both were washed several times with pentane and dried under vacuum (1.98 g, 85% yield). ^{1}H NMR ($CDCl_3$) δ : 8.77 (d, J = 12.6 Hz, 1H), 8.16 (dd, J = 7.1, 5.5 Hz, 2H), 8.05 (d, J = 7.5 Hz, 1H), 7.67 (dd, J = 8.7, 1.8 Hz, 1H), 7.56 – 7.30 (m, 8H), 4.39 (q, J = 7.1 Hz, 2H), 3.38 (d, J = 28.6 Hz, 6H), 1.47 (t, J = 7.2 Hz, 3H); ^{13}C NMR ($CDCl_3$) δ : 167.18, 166.99, 162.84, 162.75, 162.29, 162.14, 156.32, 155.68, 151.74, 151.70, 141.94, 141.83, 141.23, 140.57, 140.53, 138.30, 131.45, 131.33, 130.64, 130.35, 129.80, 129.77, 128.57, 128.49, 128.37, 128.00, 126.46, 126.40, 124.32, 123.77, 123.34, 123.09, 123.01, 122.76, 122.38, 120.86, 120.80, 119.91, 119.62, 113.81, 112.55, 108.98, 108.88, 108.60, 108.27, 37.87, 28.53, 28.01, 13.93, 13.86; HRMS (ESI MS) m/z: theor: 463.1896 found: 563.1899 ($[M]^{+\cdot}$ detected) FT-IR (ATR) 1721.56, 1657.64, 1532.47, 1451.67, 1409.62, 1350.65, 1221.52, 1154.8, 1123.83, 1079.44, 910.21, 787.23, 768.09, 748.59, 699.08

Synthesis of (*Z*/*E*)-1,3-diethyl-5-(3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-2-thioxodihydropyrimidine-4,6(1*H*,5*H*)-dione (J)



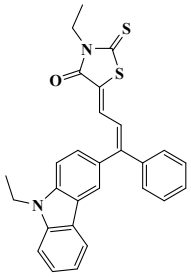
Chemical Formula: $C_{31}H_{29}N_3O_2S$
Molecular Weight: 507.6520



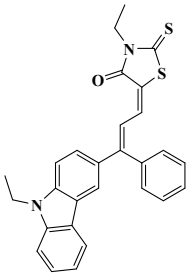
Chemical Formula: $C_{31}H_{29}N_3O_2S$
Molecular Weight: 507.6520

1,3-Diethyl-2-thioxodihydropyrimidine-4,6(1*H*,5*H*)-dione (1.0 g, 5.02 mmol, M = 200.26 g/mol) and 3,3-*bis*(4-(dimethylamino)phenyl)acrylaldehyde (1.63 g, 5.02 mmol, M = 325.41 g/mol) were dissolved in absolute ethanol (20 mL) and a few drops of piperidine were added. The solution was refluxed overnight. After cooling, the solvent was removed under reduced pressure. Addition of ether followed by pentane precipitated a blue solid that was filtered off, washed several times with pentane and dried under vacuum (1.99 g, 78% yield). ^{1}H NMR ($CDCl_3$) δ : 8.85 – 7.96 (m, 2H), 7.74 – 7.32 (m, 3H), 4.64 – 4.36 (m, 2H), 1.47 (t, *J* = 7.2 Hz, 1H), 1.30 – 1.21 (m, 2H); ^{13}C NMR ($CDCl_3$) δ : 178.99, 168.33, 168.14, 161.10, 161.02, 160.22, 160.07, 157.68, 156.93, 142.05, 141.91, 141.39, 140.59, 138.29, 131.42, 131.37, 130.88, 130.53, 129.96, 129.88, 128.68, 128.54, 128.40, 128.21, 126.54, 126.45, 124.53, 124.46, 123.44, 123.08, 123.02, 120.91, 120.81, 120.03, 119.70, 114.57, 113.30, 109.04, 108.92, 108.70, 108.33, 43.66, 43.16, 37.91, 13.86, 12.55, 12.40; HRMS (ESI MS) m/z: theor: 507.1980 found: 507.1982 ($[M]^{+\cdot}$ detected) FT-IR (ATR) : 1655.43, 1531.28, 1470.82, 1436.64, 1373.08, 1336.79, 1319.89, 1281.70, 1228.14, 1204.59, 1153.55, 1122.91, 1110.05, 1097.79, 1079.77, 998.02, 939.65, 886.64, 853.70, 832.19, 799.89, 786.09, 776.93, 744.93, 727.83, 693.81, 671.87, 593.06, 546.13

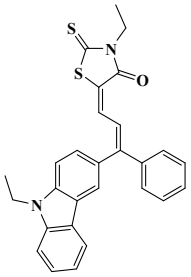
Synthesis of (Z/E)-3-ethyl-5-((Z/E)-3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-2-thioxothiazolidin-4-one (K)



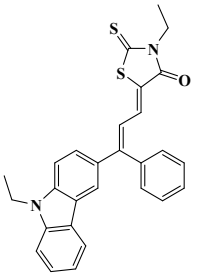
Chemical Formula: $C_{28}H_{24}N_2OS_2$
Molecular Weight: 468.6330



Chemical Formula: $C_{28}H_{24}N_2OS_2$
Molecular Weight: 468.6330



Chemical Formula: $C_{28}H_{24}N_2OS_2$
Molecular Weight: 468.6330

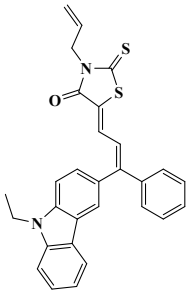


Chemical Formula: $C_{28}H_{24}N_2OS_2$
Molecular Weight: 468.6330

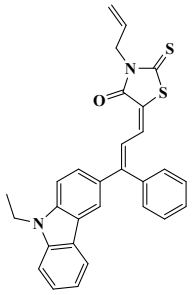
3-(9-Ethyl-9*H*-carbazol-3-yl)-3-(4-methoxyphenyl)acrylaldehyde (3.27 g, 9.21 mmol, M = 355.44 g/mol) and 3-ethylrhodanine (1.50 g, 9.21 mmol, M = 161.25 g/mol) were suspended in ethanol (30 mL) and the flask was introduced into a preheated bath at 100°C. A few drops of piperidine were added and the solution was refluxed for 20 min. During reflux, a purple precipitate formed. It was filtered off, washed several times with ethanol and dried under vacuum. (3.54 g, 82% yield). ^{1}H NMR ($CDCl_3$) δ : 8.00 – 7.87 (m, J = 20.1 Hz, 2H), 7.48 – 7.22 (m, 10H), 7.18 – 7.14 (m, 1H), 6.61 (dd, J = 43.3, 12.1 Hz, 1H), 4.39 – 4.28 (m, 2H), 4.11 – 4.02 (m, 2H), 1.44 – 1.36 (m, 3H), 1.21 – 1.15 (m, 3H); ^{13}C NMR ($CDCl_3$) δ : 192.33, 166.85, 166.79, 156.81, 141.96, 140.93, 140.52, 140.47, 138.72, 131.95, 131.73, 130.84, 129.63, 129.10, 128.79, 128.67,

128.53, 126.61, 126.33, 126.26, 124.87, 123.83, 123.21, 123.07, 122.90, 122.71, 121.92, 121.30, 120.75, 120.71, 119.62, 119.40, 108.87, 108.79, 108.51, 108.40, 39.64, 37.78, 13.92, 13.84, 12.24; HRMS (ESI MS) m/z: theor: 468.1330 found: 468.1333 ($[M]^{+\cdot}$ detected) FT-IR (ATR) : 1697.79, 1560.24, 1470.87, 1320.38, 1230.02, 1122.56, 873.33, 806.06, 768.35, 738.63, 696.83, 508.67

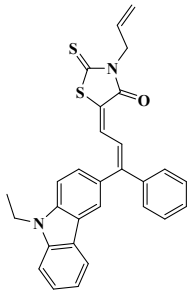
Synthesis of (Z/E)-3-allyl-5-((Z/E)-3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-2-thioxo-thiazolidin-4-one (L)



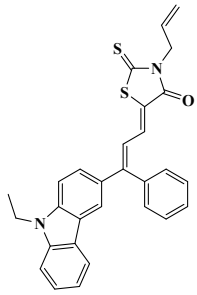
Chemical Formula: $C_{29}H_{24}N_2OS_2$
Molecular Weight: 480.6440



Chemical Formula: $C_{29}H_{24}N_2OS_2$
Molecular Weight: 480.6440



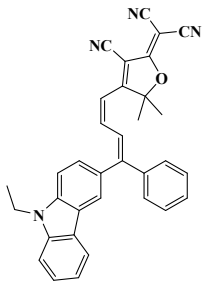
Chemical Formula: $C_{29}H_{24}N_2OS_2$
Molecular Weight: 480.6440



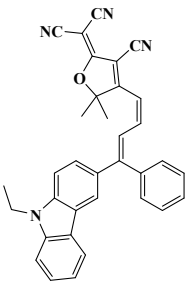
Chemical Formula: $C_{29}H_{24}N_2OS_2$
Molecular Weight: 480.6440

3-3-(9-Ethyl-9*H*-carbazol-3-yl)-3-(4-methoxyphenyl)acrylaldehyde (3.27 g, 9.21 mmol, M = 355.44 g/mol) and 3-allylrhodanine (1.09 g, 9.21 mmol, M = 173.26 g/mol) were suspended in ethanol (30 mL) and the flask was introduced into a preheated bath at 100°C. A few drops of piperidine were added and the solution was refluxed for 20 min. During reflux, a purple precipitate formed. It was filtered off, washed several times with ethanol and dried under vacuum (3.72 g, 84% yield). ^{1}H NMR ($CDCl_3$) δ : 8.02 (t, *J* = 16.3 Hz, 2H), 7.57 – 7.30 (m, 11H), 6.69 (dd, *J* = 33.2, 12.1 Hz, 1H), 5.93 – 5.75 (m, 1H), 5.33 – 5.15 (m, 2H), 4.69 (t, *J* = 6.5 Hz, 2H), 4.41 (td, *J* = 14.4, 7.2 Hz, 2H), 1.46 (t, *J* = 7.3 Hz, 3H); ^{13}C NMR ($CDCl_3$) δ : 192.13, 166.65, 157.03, 141.93, 140.47, 132.24, 132.02, 130.81, 129.76, 129.66, 129.18, 129.10, 128.77, 128.64, 128.52, 128.42, 126.62, 126.33, 126.26, 124.55, 123.17, 123.09, 122.71, 121.85, 121.31, 120.76, 120.70, 119.63, 119.40, 119.15, 119.09, 108.78, 108.52, 108.39, 46.29, 37.78, 13.90, 13.82; HRMS (ESI MS) m/z: theor: 480.1330 found: 480.1331 ($[M]^{+\cdot}$ detected) FT-IR (ATR): 1698.37, 1558.49, 1471.32, 1366.69, 1313.21, 1205.16, 1125.34, 907.58, 805.93, 769.74, 739.38, 696.15

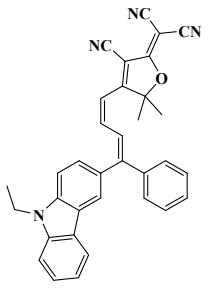
Synthesis of 2-(3-cyano-4-((1Z/*E*,3Z/*E*)-4-(9-ethyl-9*H*-carbazol-3-yl)-4-phenylbuta-1,3-dien-1-yl)-5,5-dimethylfuran-2(5*H*)-ylidene)malononitrile (M)



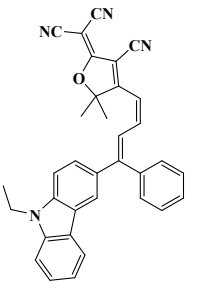
Chemical Formula: $C_{34}H_{26}N_4O$
Molecular Weight: 506.6090



Chemical Formula: $C_{34}H_{26}N_4O$
Molecular Weight: 506.6090



Chemical Formula: $C_{34}H_{26}N_4O$
Molecular Weight: 506.6090

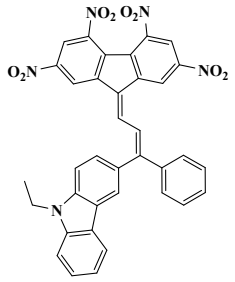


Chemical Formula: $C_{34}H_{26}N_4O$
Molecular Weight: 506.6090

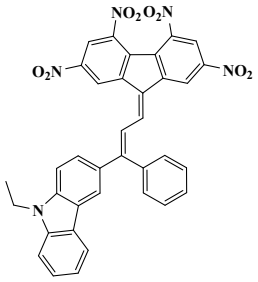
2-(3-Cyano-4,5,5-trimethylfuran-2(5*H*)-ylidene)malononitrile (1 g, 5.02 mmol, M = 199.21 g/mol) 3,3-*bis*(4-(dimethylamino)phenyl)acrylaldehyde (1.63 g, 5.02 mmol, M = 325.41 g/mol) were dissolved in absolute ethanol (20 mL) and a few drops of piperidine were added. The solution was refluxed

overnight. After cooling, the solvent was removed under reduced pressure. Addition of ether followed by pentane precipitated a blue solid that was filtered off, washed several times with pentane and dried under vacuum (2.26 g, 89% yield). ^{1}H NMR ($CDCl_3$) δ : 1.47 (t, 3H, J = 7.2 Hz), 1.54 (s, 6H), 4.40 (q, 2H, J = 7.2 Hz), 6.66 (d, 1H, J = 15.3 Hz), 7.18 (d, 1H, J = 11.5 Hz), 7.25-7.35 (m, 5H), 7.37-7.46 (m, 2H), 7.50-7.56 (m, 4H), 8.04 (d, 1H, J = 7.7 Hz), 8.10 (d, 1H, J = 1.4 Hz); ^{13}C NMR ($CDCl_3$) δ : 13.8, 26.4, 37.9, 56.2, 96.9, 97.2, 108.8, 109.0, 110.5, 111.3, 112.2, 117.1, 119.9, 120.7, 121.8, 122.9, 123.5, 124.7, 126.6, 126.9, 128.6, 129.5, 129.6, 130.5, 130.8, 138.6, 140.6, 141.5, 147.2, 159.5, 174.1, 175.6; HRMS (ESI MS) m/z: theor: 506.2107 found: 506.2110 ($[M]^{+\cdot}$ Detected) FT-IR (ATR): 2218.72, 1537.06, 1481.20, 1391.75, 1364.86, 1343.28, 1265.75, 1233.06, 1211.25, 1182.16, 1154.87, 1122.83, 976.07, 939.58, 891.03, 852.02, 805.82, 766.31, 752.08, 733.66, 719.45, 702.99, 649.62, 581.87, 559.35, 512.54

Synthesis of (Z/E)-9-ethyl-3-(1-phenyl-3-(2,4,5,7-tetranitro-9*H*-fluoren-9-ylidene)prop-1-en-1-yl)-9*H*-carbazole (N)



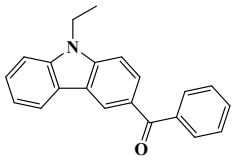
Chemical Formula: $C_{36}H_{23}N_5O_8$
Molecular Weight: 653.6070



Chemical Formula: $C_{36}H_{23}N_5O_8$
Molecular Weight: 653.6070

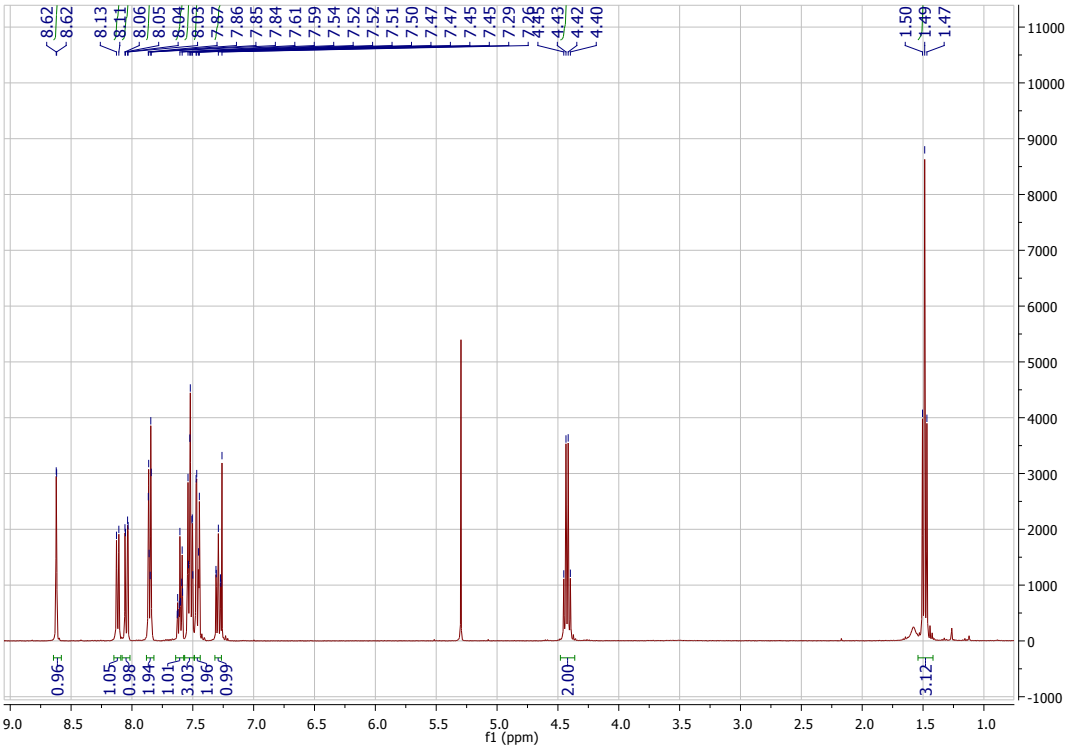
3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylacrylaldehyde (941 mg, 2.89 mmol, M = 325.41 g/mol) was added to a solution of 2,4,5,7-tetranitro-9*H*-fluorene (1 g, 2.89 mmol, M = 346.21 g/mol) in DMF (10 mL) and the mixture was stirred at room temperature for two hours. The solvent was removed under reduced pressure. Dissolution in ether followed by addition of pentane precipitated a deep blue solid that was filtered off, washed several times with pentane and dried under vacuum (1.64 g, 87% yield). ^{1}H NMR ($CDCl_3$) δ : 1.51-1.58 (m, 4.4H), 4.43-4.54 (m, 3.6H), 7.48-7.61 (m, 11H), 7.64-7.72 (m, 4.2H), 7.83-7.93 (m, 2H), 8.01-8.14 (m, 6.9H), 8.24 (d, 1H, J = 1.8 Hz), 8.60-8.61 (m, 1.6H), 8.77-8.80 (m, 1.7H), 8.90-8.92 (m, 1.7H), 9.35 (d, 0.7H, J = 1.8 Hz), 9.44 (d, 1H, J = 1.8 Hz); Anal. Calc. for $C_{36}H_{23}N_5O_8$: C, 66.2; H, 3.5; O, 19.6; Found: C, 66.2; H, 3.4; O, 19.7 %; ; HRMS (ESI MS) m/z: theor: 653.1547 found: 653.1542 ($[M]^{+\cdot}$ Detected) FT-IR (ATR): 1671.50, 1579.40, 1527.27, 1451.39, 1408.14, 1336.08, 1305.95, 1260.71, 1226.82, 1152.63, 1123.21, 1088.76, 897.18, 828.96, 769.67, 727.45, 719.99, 700.75

(9-Ethyl-9*H*-carbazol-3-yl)(phenyl)methanone

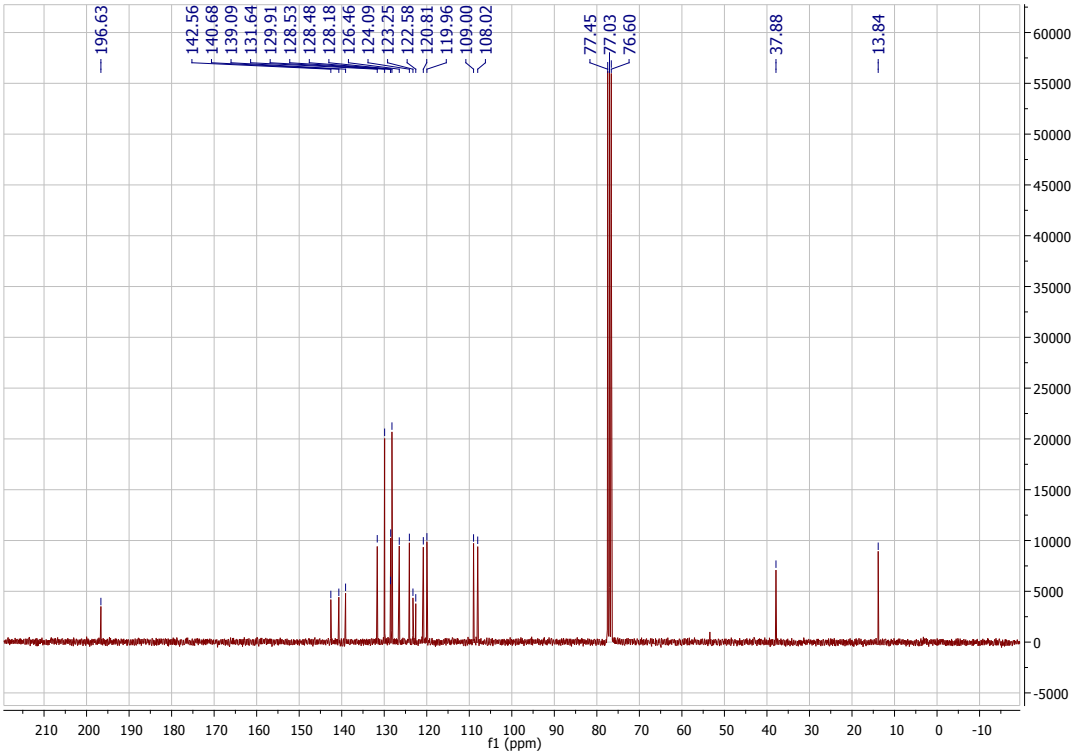


Chemical Formula: $C_{21}H_{17}NO$
Molecular Weight: 299.3730

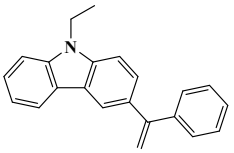
^{1}H NMR spectrum in $CDCl_3$



^{13}C NMR spectrum in $CDCl_3$

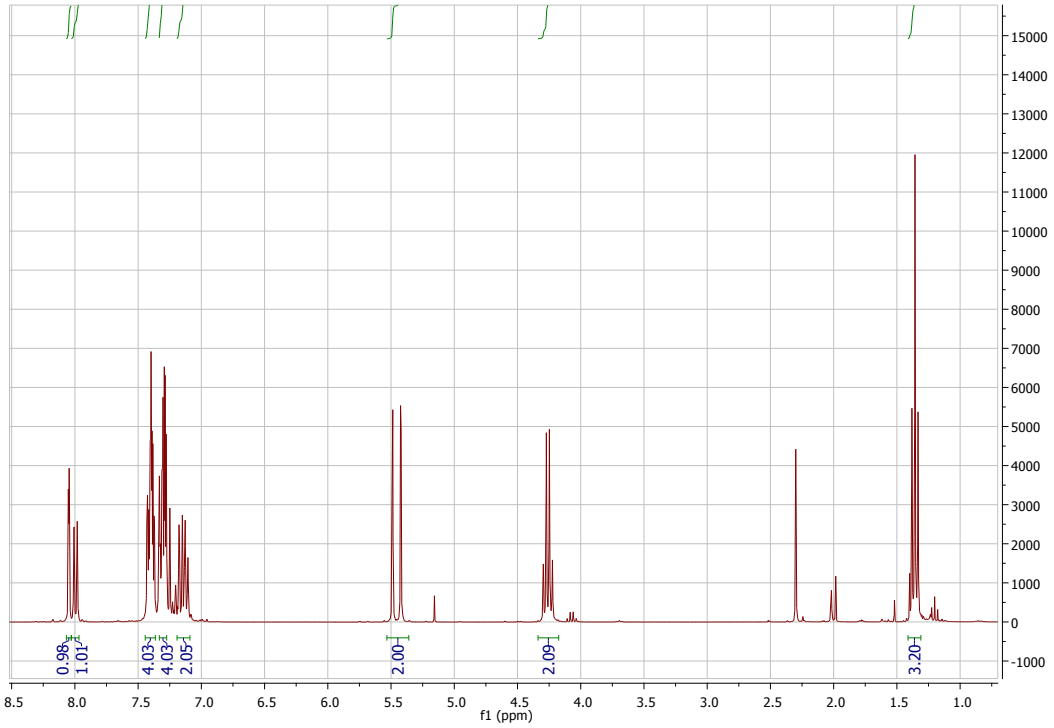


9-Ethyl-3-(1-phenylvinyl)-9*H*-carbazole

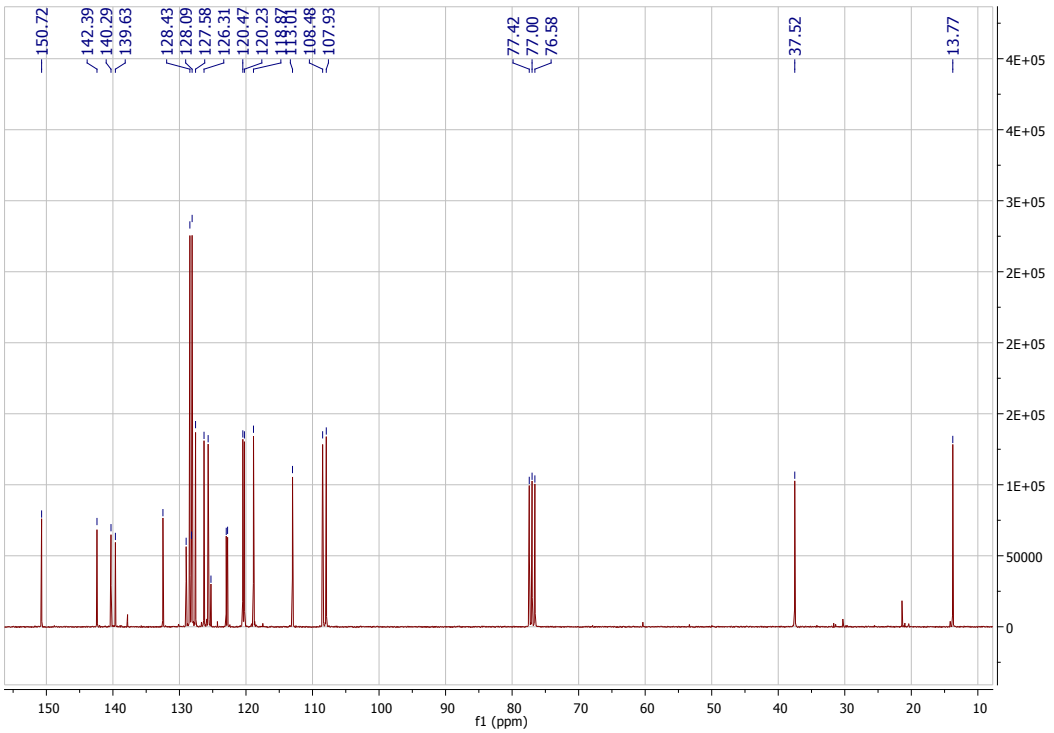


Chemical Formula: $C_{22}H_{19}N$
Molecular Weight: 297.4010

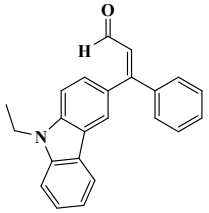
^{1}H NMR spectrum in $CDCl_3$



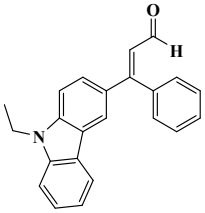
^{13}C NMR spectrum in $CDCl_3$



(Z/E)-3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylacrylaldehyde

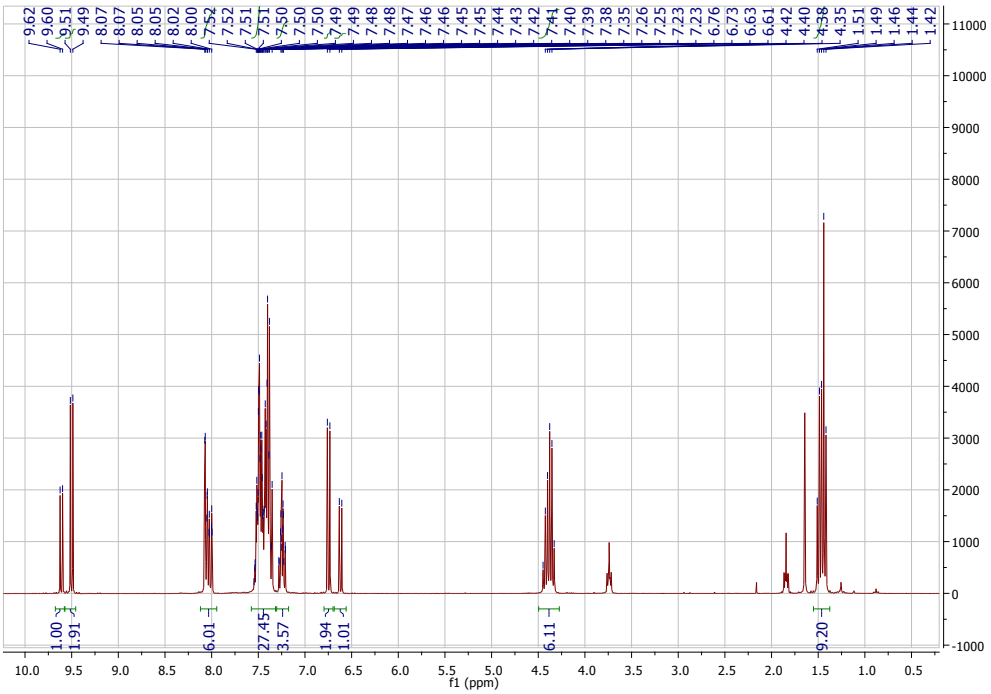


Chemical Formula: $C_{23}H_{19}NO$
Molecular Weight: 325.4110

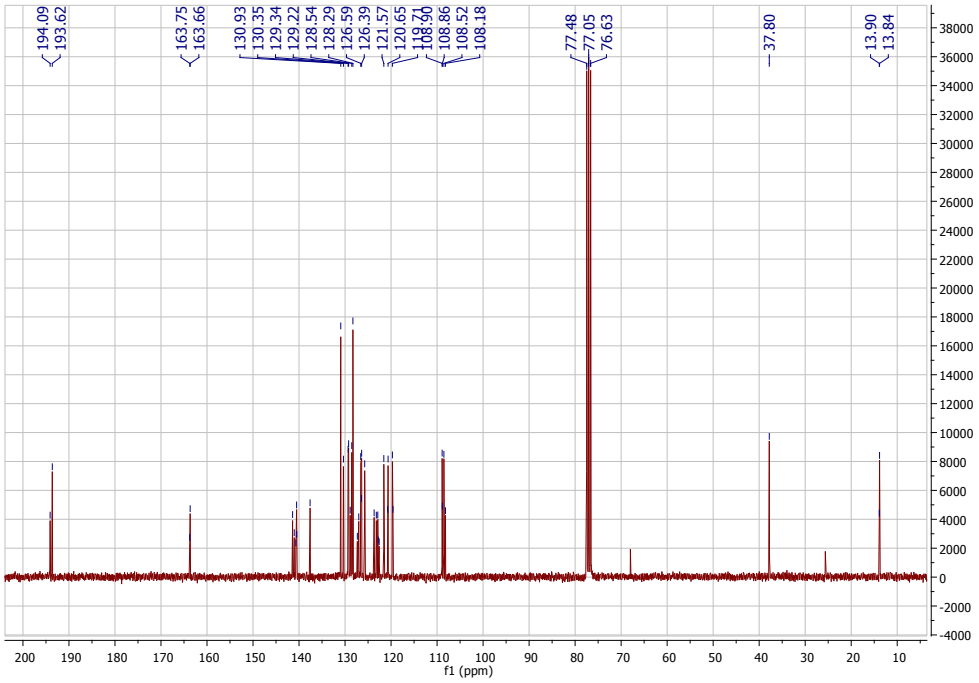


Chemical Formula: $C_{23}H_{19}NO$
Molecular Weight: 325.4110

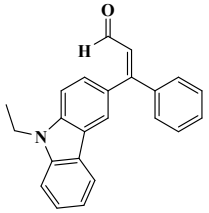
^{1}H NMR spectrum in $CDCl_3$



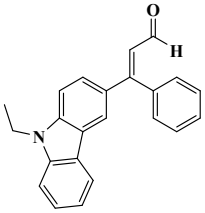
^{13}C NMR spectrum in $CDCl_3$



(3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylacrylaldehyde (separated isomer)

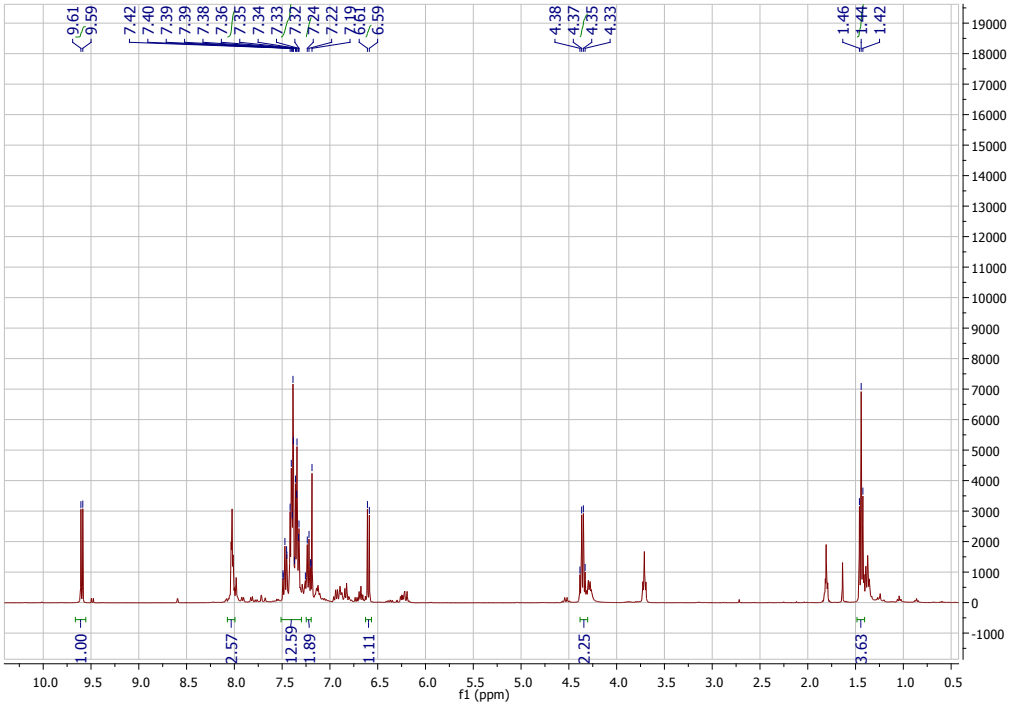


Chemical Formula: $C_{23}H_{19}NO$
Molecular Weight: 325.4110

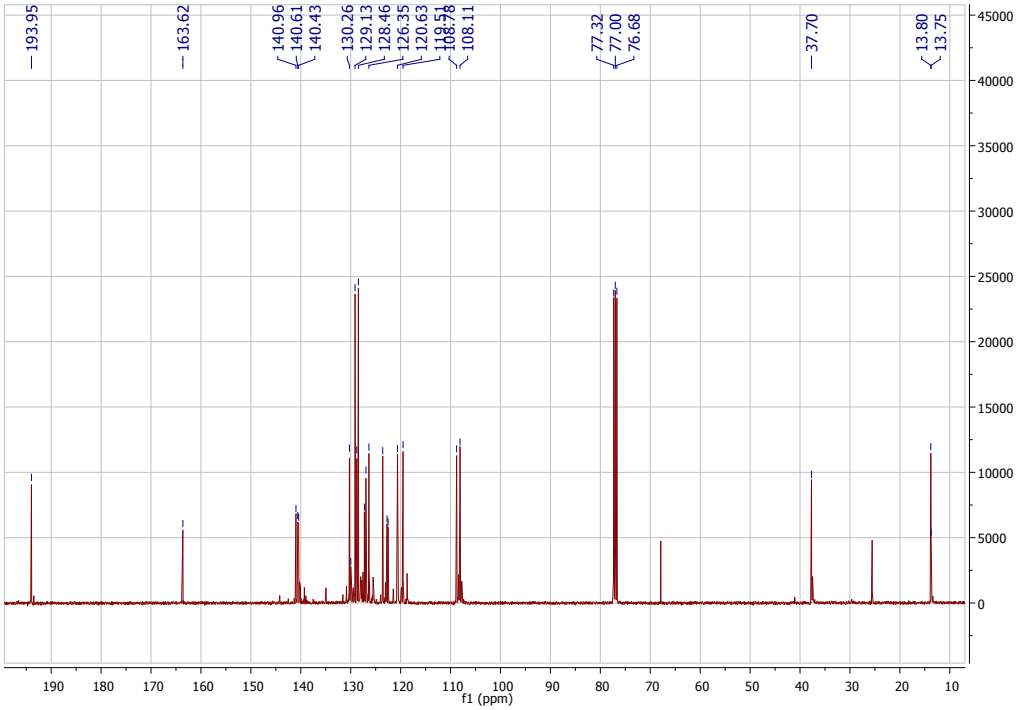


Chemical Formula: $C_{23}H_{19}NO$
Molecular Weight: 325.4110

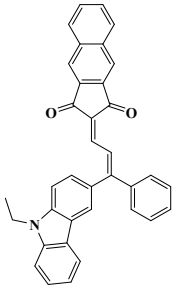
^{1}H NMR spectrum in $CDCl_3$



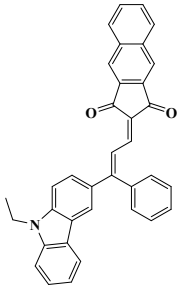
^{13}C NMR spectrum in $CDCl_3$



(Z/E)-2-(3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1*H*-cyclopenta[*b*]naphthalene -1,3(2*H*)-dione (A)

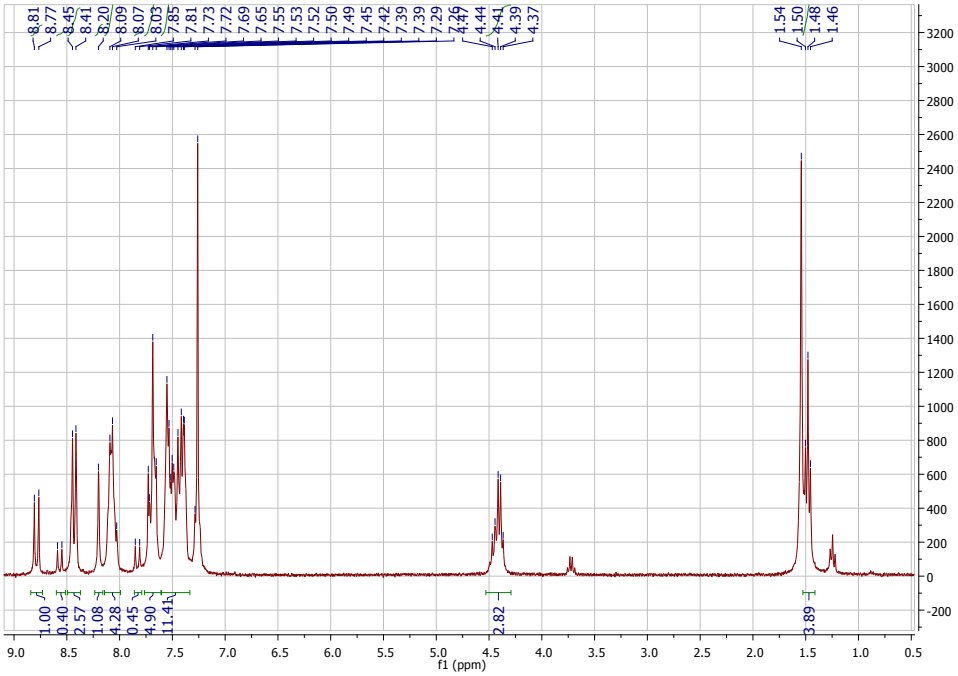


Chemical Formula: $C_{36}H_{25}NO_2$
Molecular Weight: 503.6010

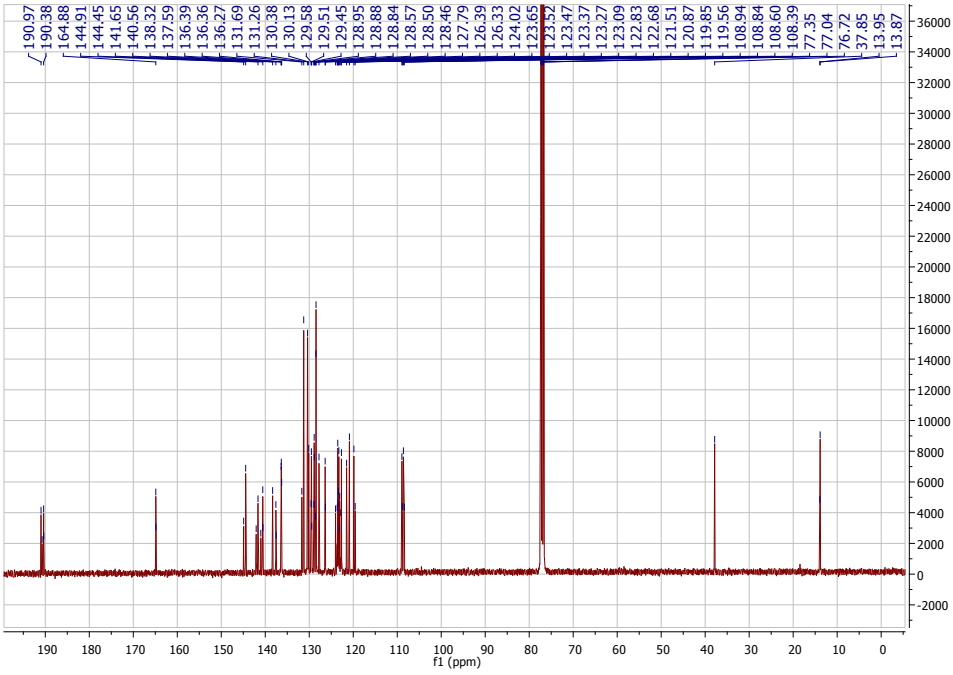


Chemical Formula: $C_{36}H_{25}NO_2$
Molecular Weight: 503.6010

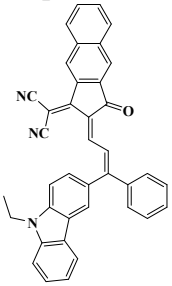
^{1}H NMR spectrum in $CDCl_3$



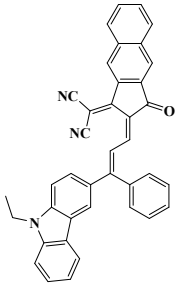
^{13}C NMR spectrum in $CDCl_3$



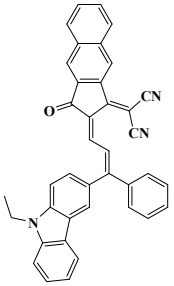
2-((*Z*/*E*)-2-((*Z*/*E*)-3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-3-oxo-2,3-dihydro-1*H*-cyclopenta[*b*]naphthalen-1-ylidene)malononitrile (B)



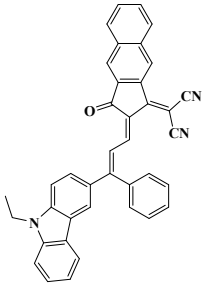
Chemical Formula: $C_{39}H_{25}N_3O$
Molecular Weight: 551.6490



Chemical Formula: $C_{39}H_{25}N_3O$
Molecular Weight: 551.6490

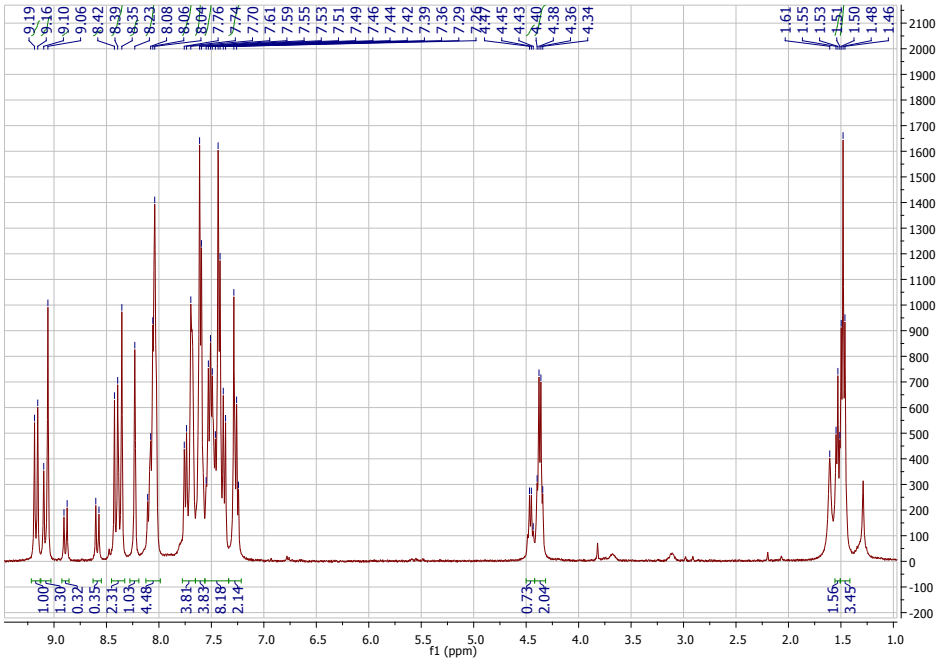


Chemical Formula: $C_{39}H_{25}N_3O$
Molecular Weight: 551.6490

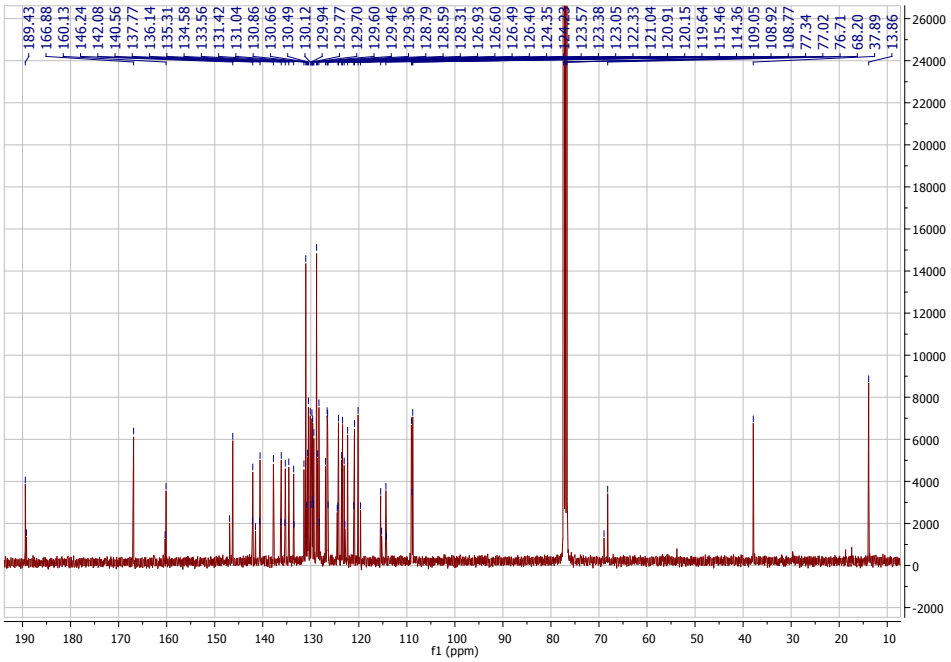


Chemical Formula: $C_{39}H_{25}N_3O$
Molecular Weight: 551.6490

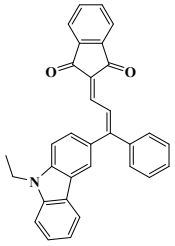
^{1}H NMR spectrum in $CDCl_3$



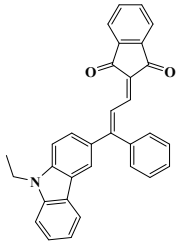
^{13}C NMR spectrum in $CDCl_3$



(*Z*/*E*)-2-(3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1*H*-indene-1,3(2*H*)-dione (C/D)

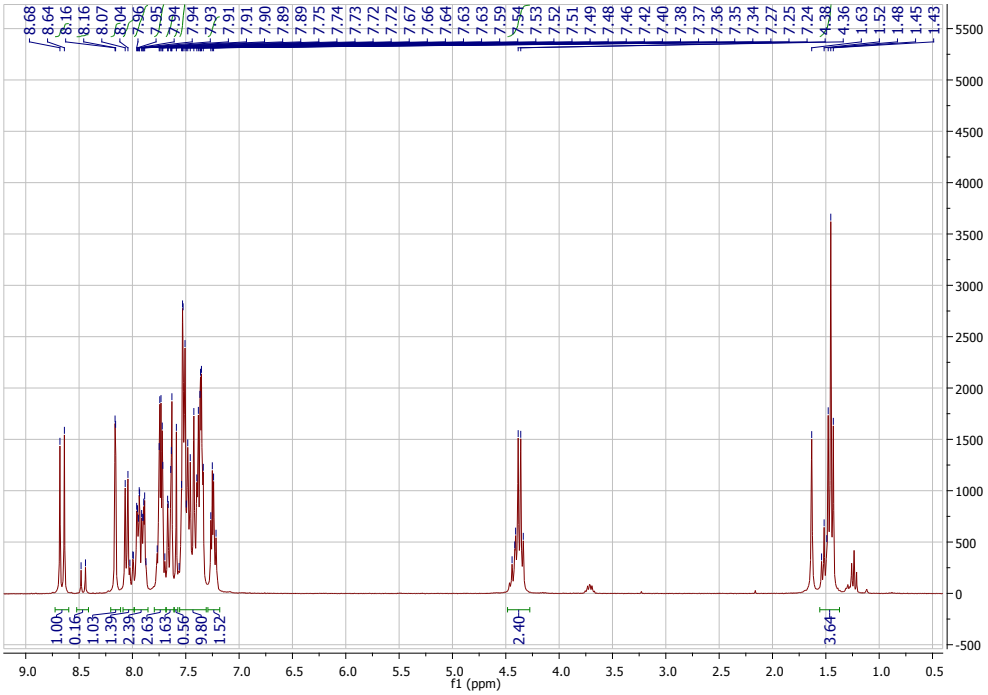


Chemical Formula: $C_{32}H_{23}NO_2$
Molecular Weight: 453.5410

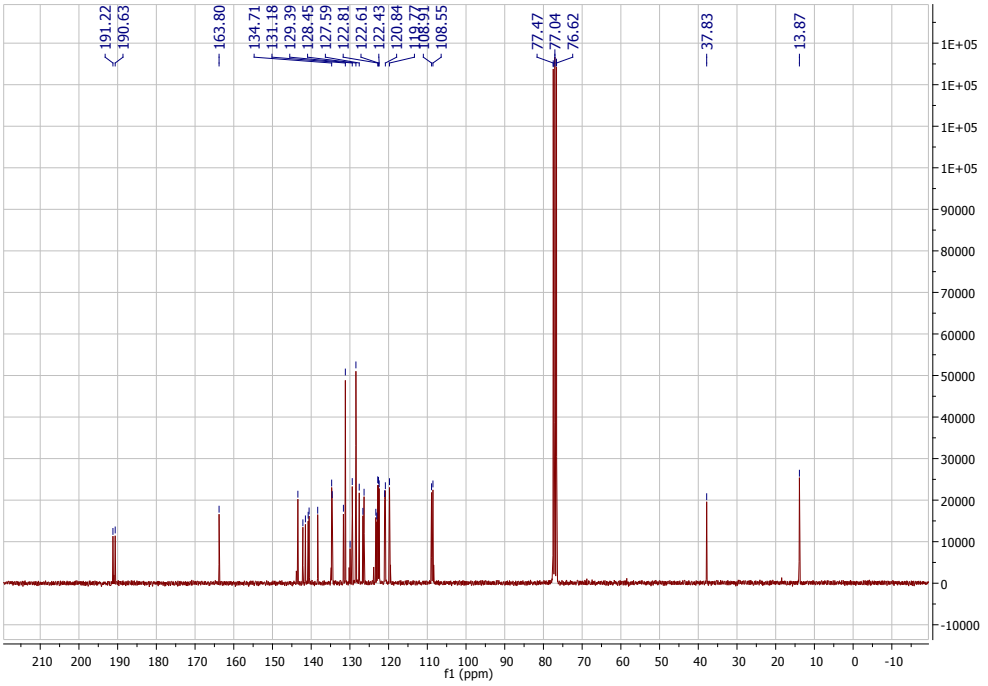


Chemical Formula: $C_{32}H_{23}NO_2$
Molecular Weight: 453.5410

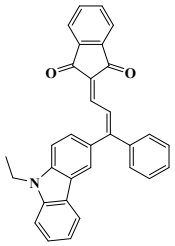
^{1}H NMR spectrum in $CDCl_3$



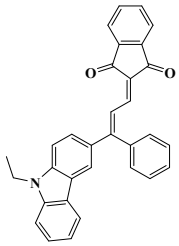
^{13}C NMR spectrum in $CDCl_3$



2-(3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1*H*-indene-1,3(2*H*)-dione (C/D) (separated isomer)

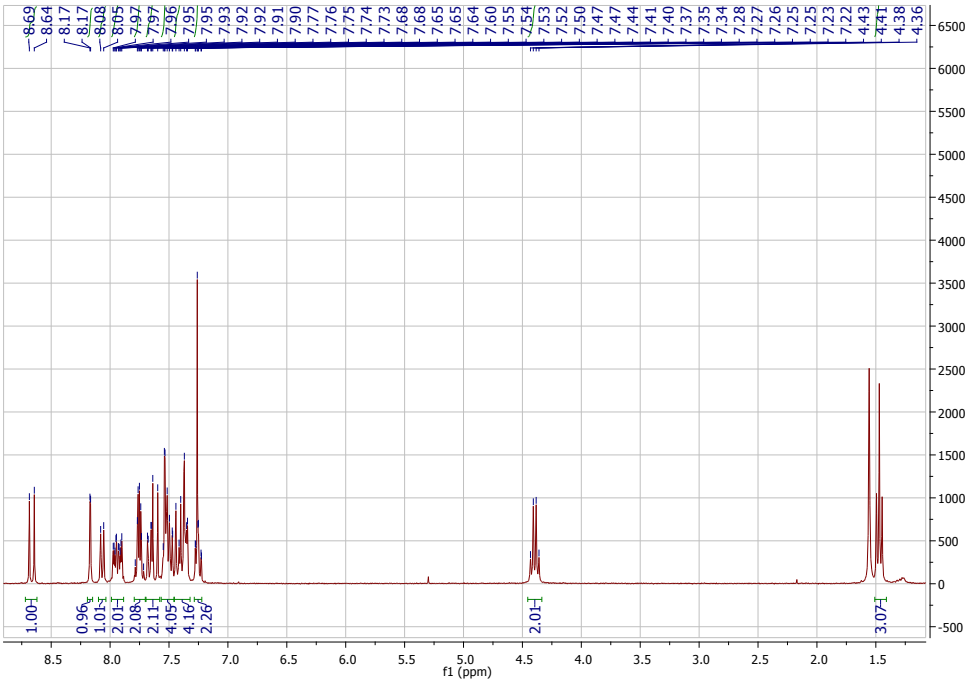


Chemical Formula: $C_{32}H_{23}NO_2$
Molecular Weight: 453.5410

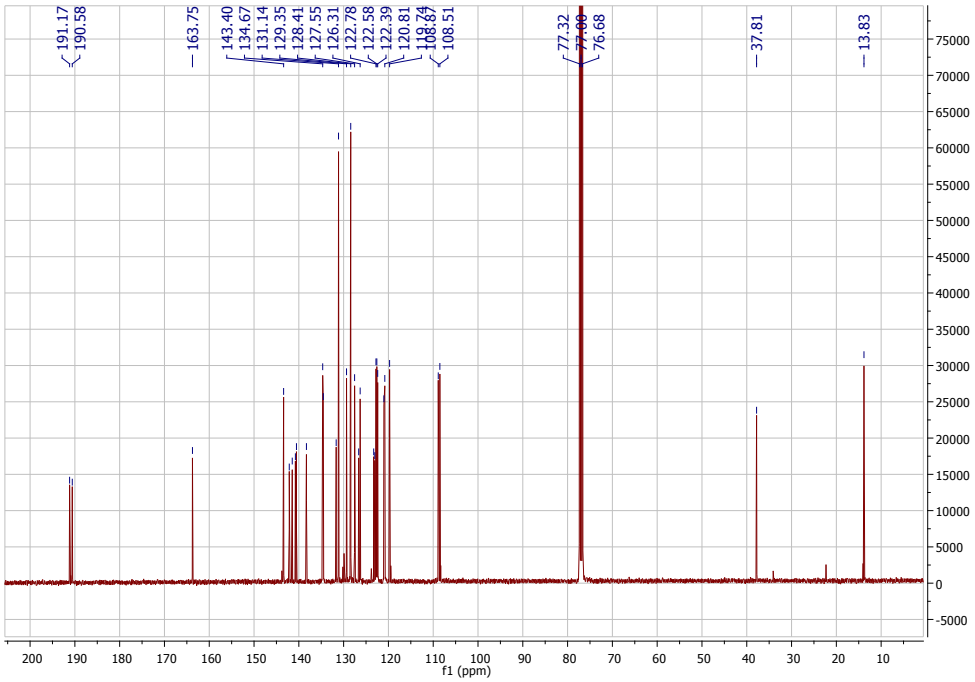


Chemical Formula: $C_{32}H_{23}NO_2$
Molecular Weight: 453.5410

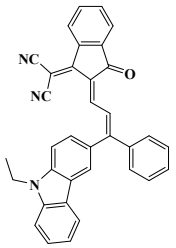
^{1}H NMR spectrum in $CDCl_3$



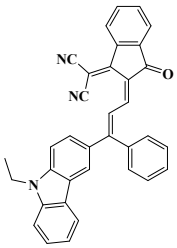
^{13}C NMR spectrum in $CDCl_3$



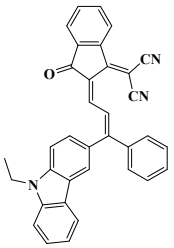
2-((*Z*/*E*)-2-((*Z*/*E*)-3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-3-oxo-2,3-dihydro-1*H*-inden-1-ylidene)malononitrile (E)



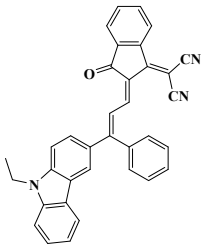
Chemical Formula: $C_{35}H_{23}N_3O$
Molecular Weight: 501.5890



Chemical Formula: $C_{35}H_{23}N_3O$
Molecular Weight: 501.5890

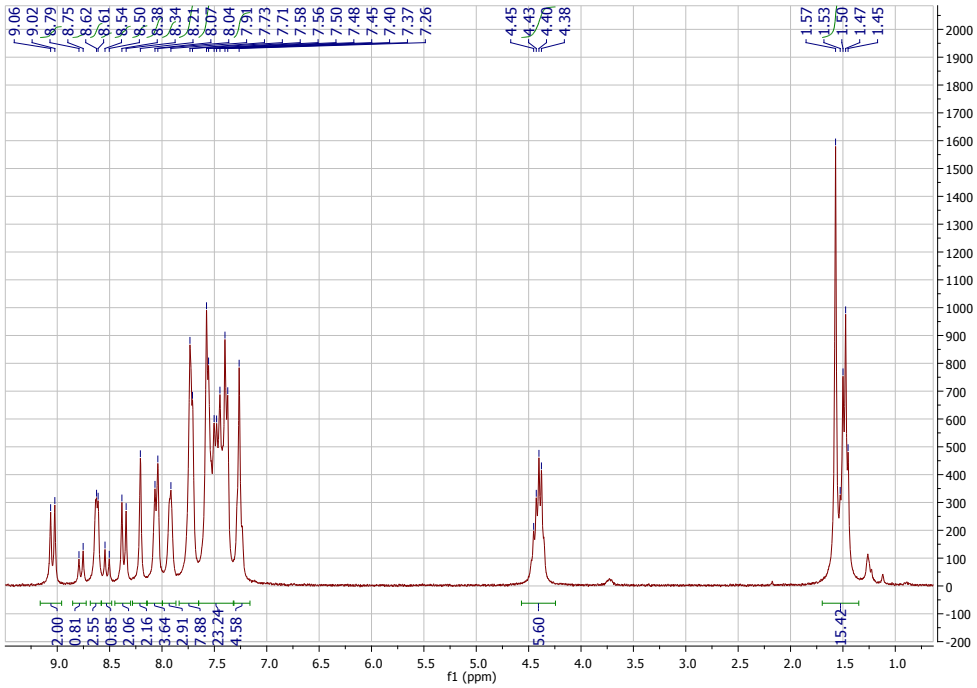


Chemical Formula: $C_{35}H_{23}N_3O$
Molecular Weight: 501.5890

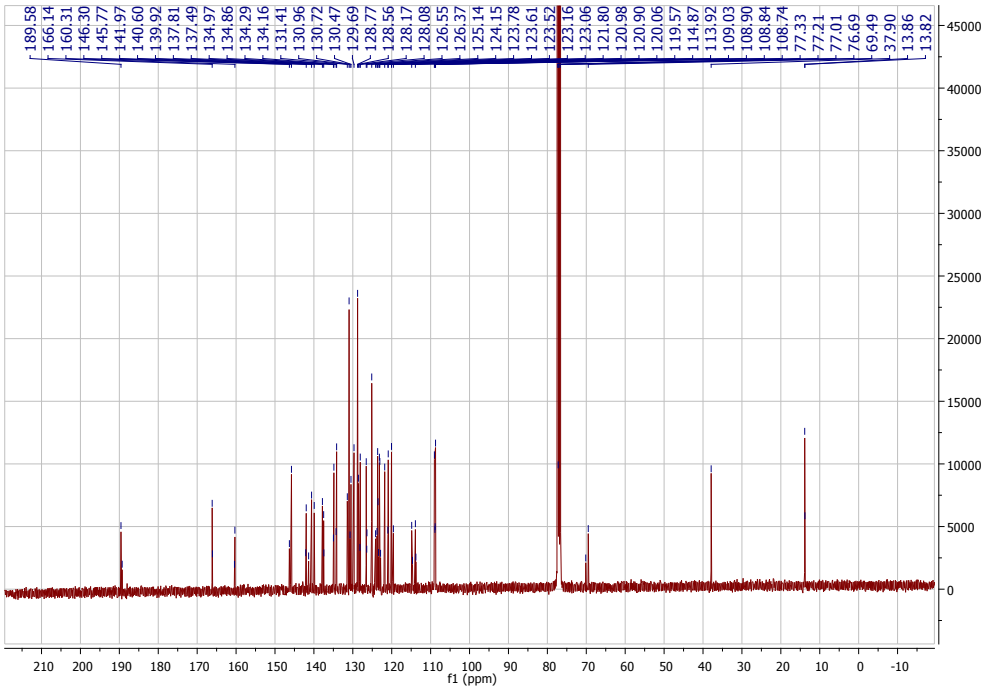


Chemical Formula: $C_{35}H_{23}N_3O$
Molecular Weight: 501.5890

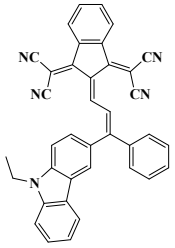
^{1}H NMR spectrum in $CDCl_3$



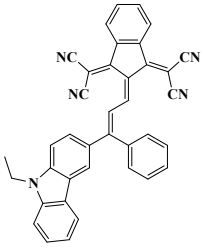
^{13}C NMR spectrum in $CDCl_3$



(*Z*/*E*)-2,2'-(2-(3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1*H*-indene-1,3(2*H*)-diylidene)dimalononitrile (F)

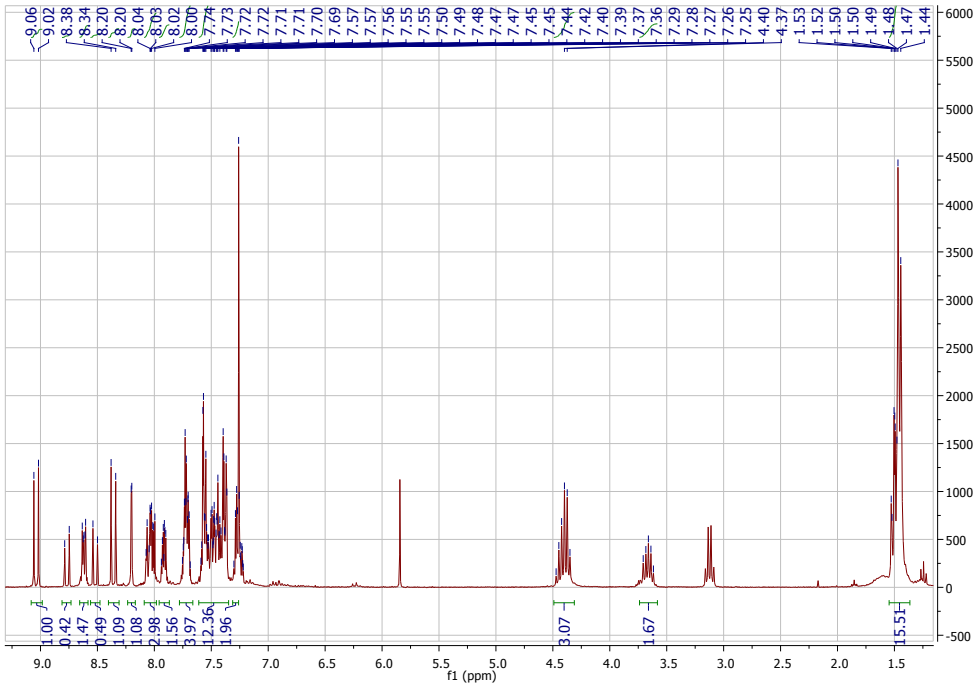


Chemical Formula: $C_{38}H_{23}N_5$
Molecular Weight: 549.6370

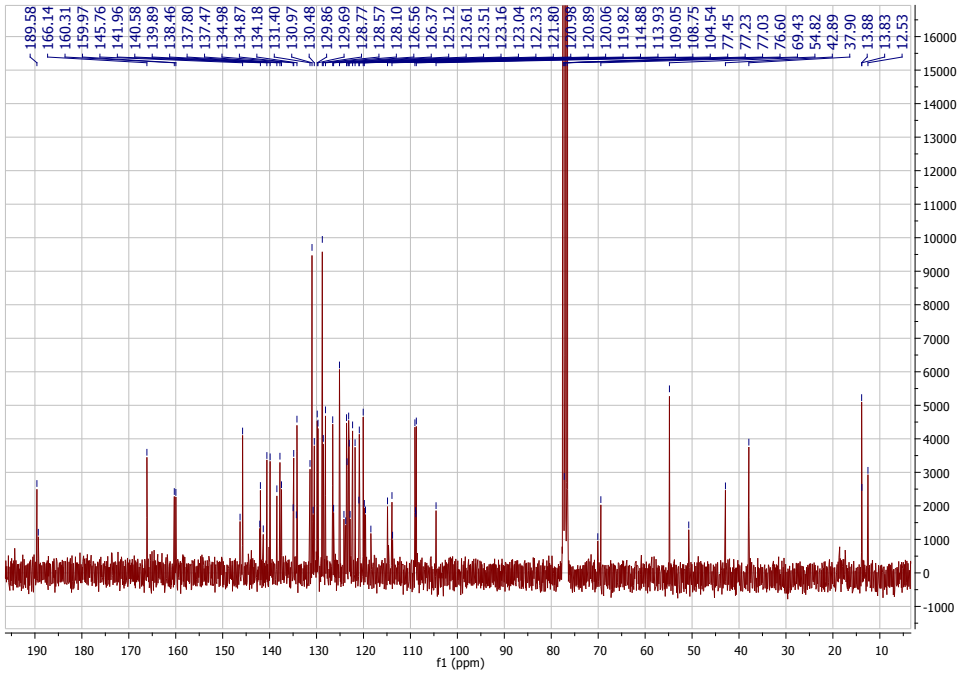


Chemical Formula: $C_{38}H_{23}N_5$
Molecular Weight: 549.6370

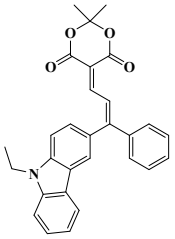
^{1}H NMR spectrum in $CDCl_3$



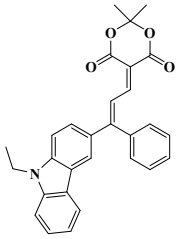
^{13}C NMR spectrum in $CDCl_3$



(Z/E)-5-(3-(9-Ethyl-9H-carbazol-3-yl)-3-phenylallylidene)-2,2-dimethyl-1,3-dioxane-4,6-dione (G)

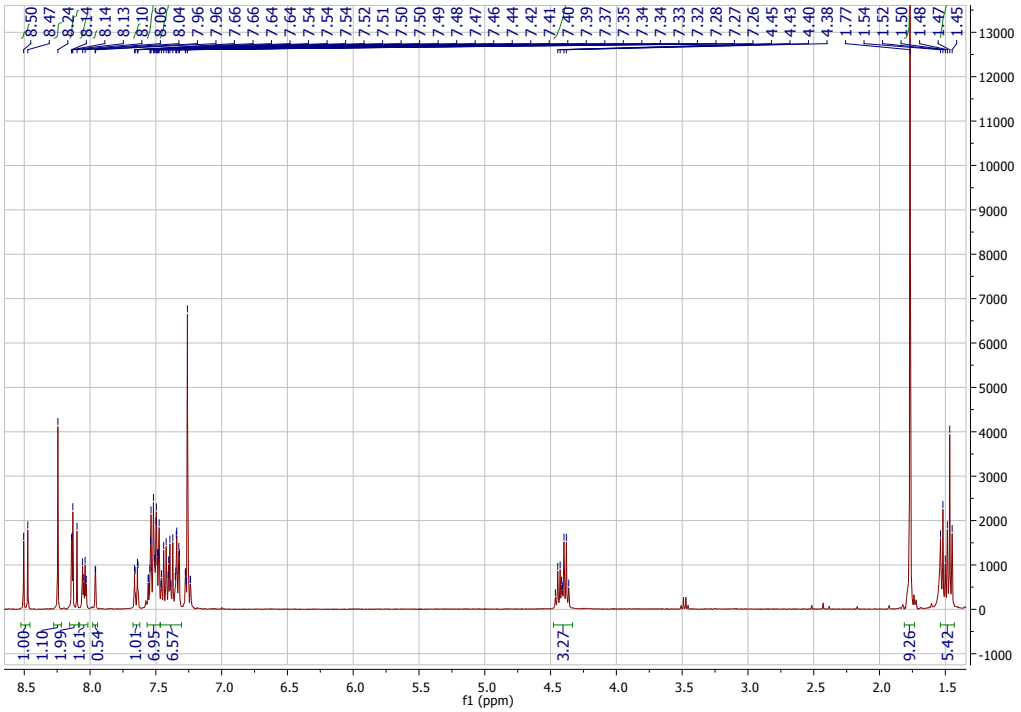


Chemical Formula: $C_{29}H_{25}NO_4$
Molecular Weight: 451.5220

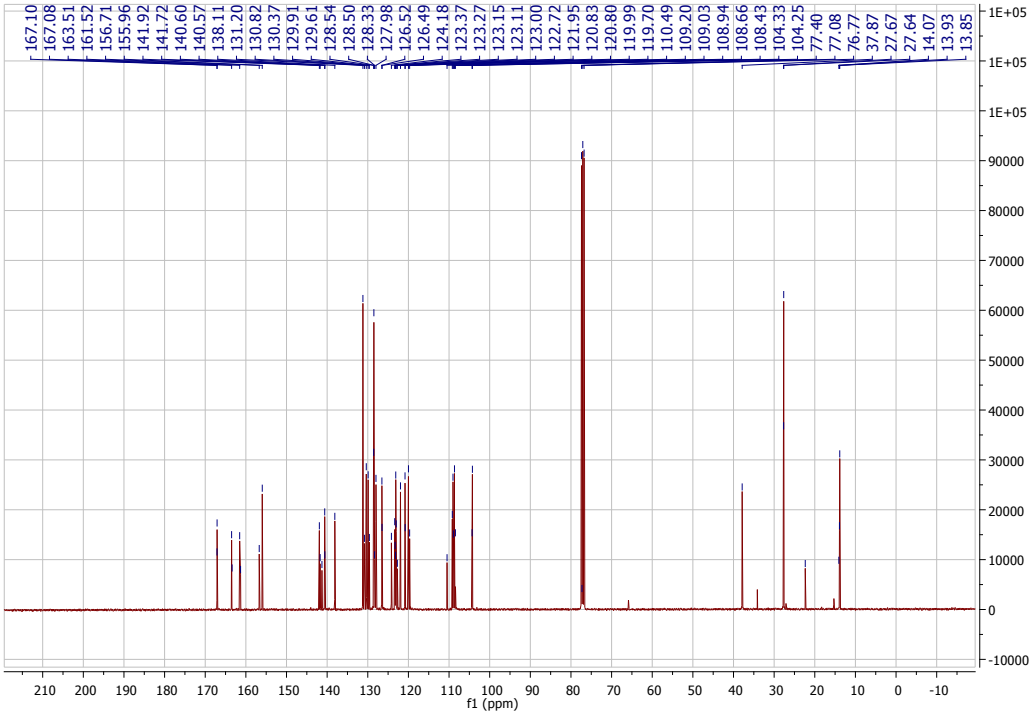


Chemical Formula: $C_{29}H_{25}NO_4$
Molecular Weight: 451.5220

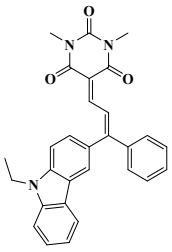
^{1}H NMR spectrum in $CDCl_3$



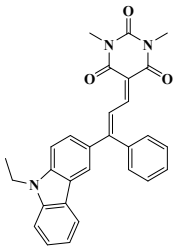
^{13}C NMR spectrum in $CDCl_3$



(*Z*/*E*)-5-(3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1,3-dimethylpyrimidine-2,4,6(1*H*,3*H*,5*H*)-trione (H/I)

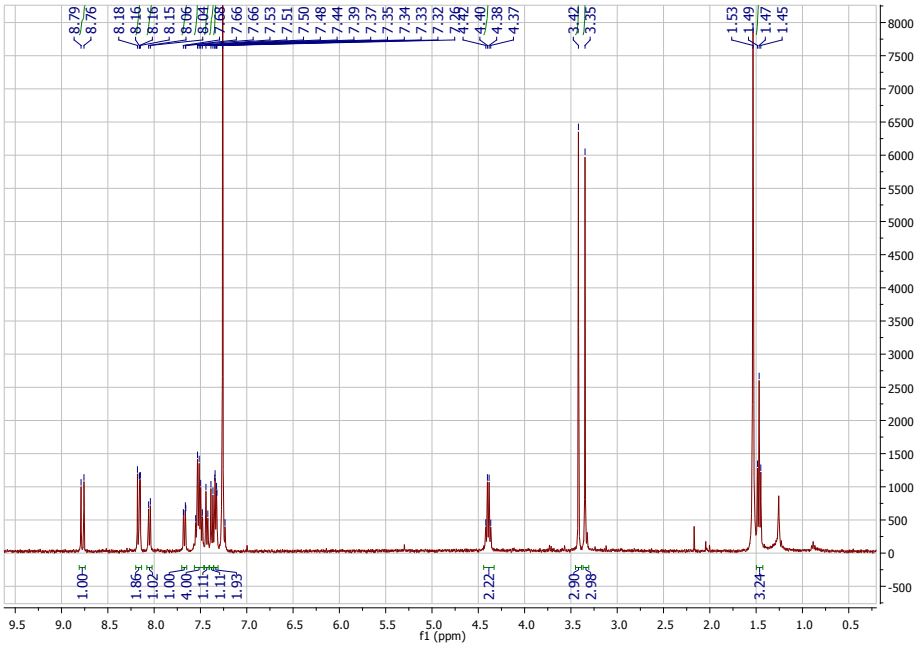


Chemical Formula: $C_{29}H_{25}N_3O_3$
Molecular Weight: 463.5370

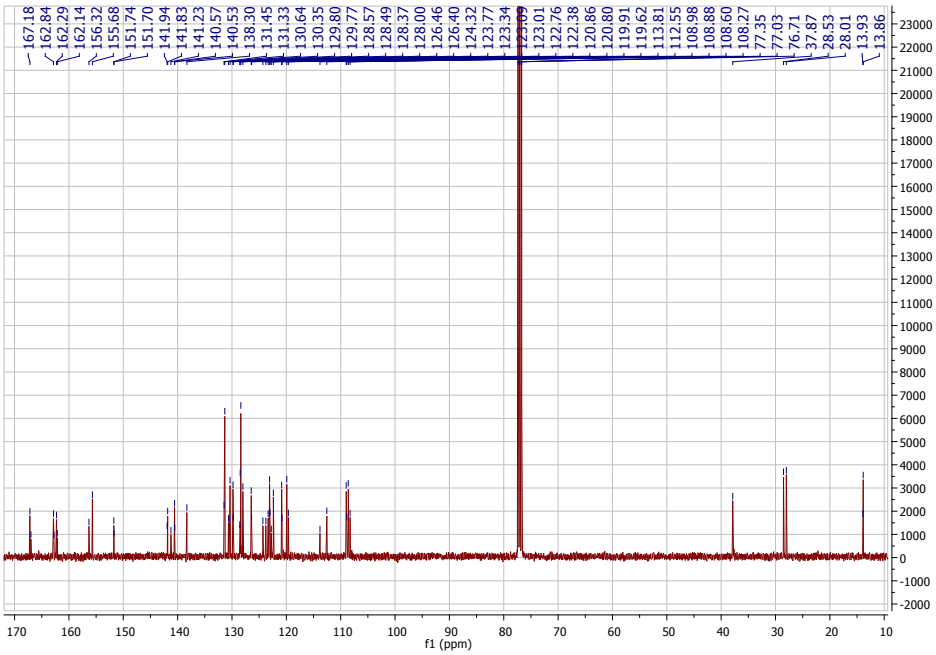


Chemical Formula: $C_{29}H_{25}N_3O_3$
Molecular Weight: 463.5370

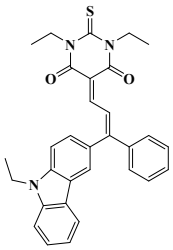
^{1}H NMR spectrum in $CDCl_3$



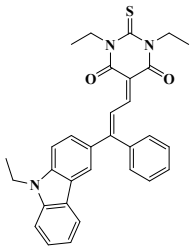
^{13}C NMR spectrum in $CDCl_3$



(*Z*/*E*)-1,3-Diethyl-5-(3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-2-thioxodihydro-pyrimidine-4,6(1*H*,5*H*)-dione (J)

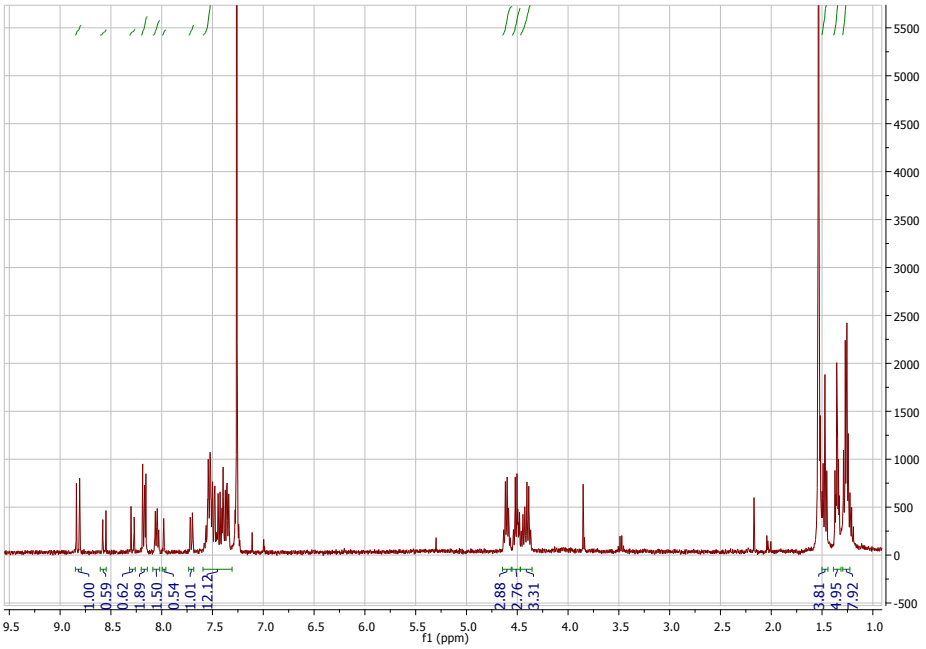


Chemical Formula: $C_{31}H_{29}N_3O_2S$
Molecular Weight: 507.6520

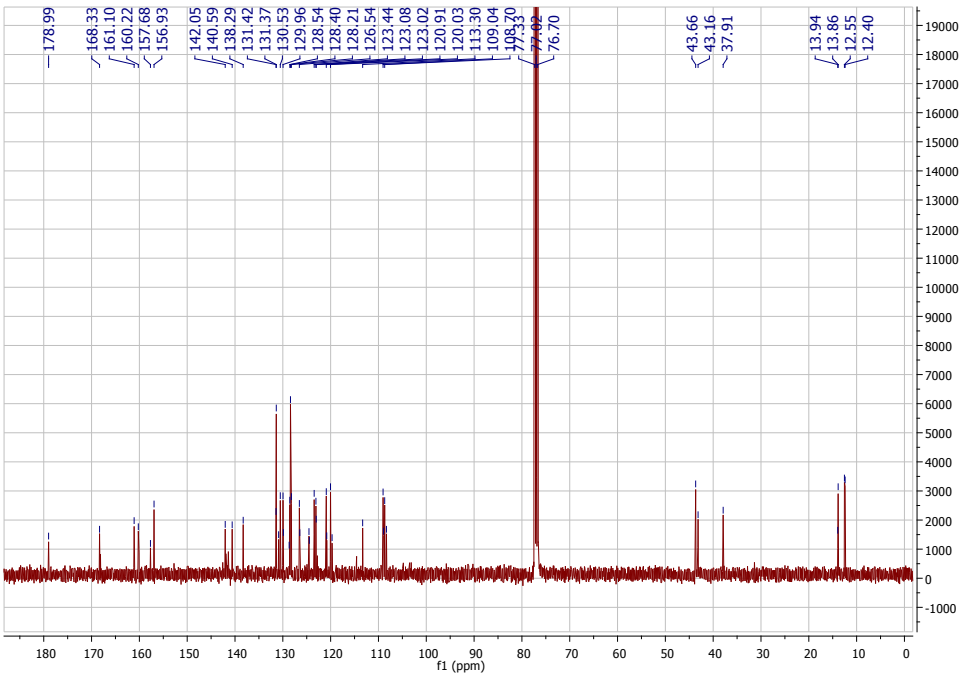


Chemical Formula: $C_{31}H_{29}N_3O_2S$
Molecular Weight: 507.6520

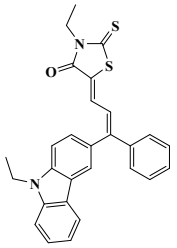
^{1}H NMR spectrum in $CDCl_3$



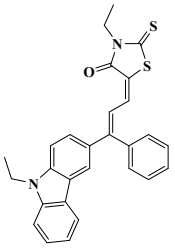
^{13}C NMR spectrum in $CDCl_3$



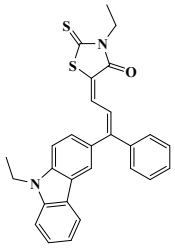
(Z/E)-3-Ethyl-5-((Z/E)-3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-2-thioxo-thiazolidin-4-one (K)



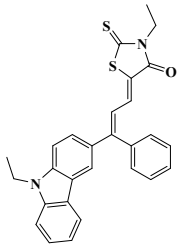
Chemical Formula: $C_{28}H_{24}N_2OS_2$
Molecular Weight: 468.6330



Chemical Formula: $C_{28}H_{24}N_2OS_2$
Molecular Weight: 468.6330

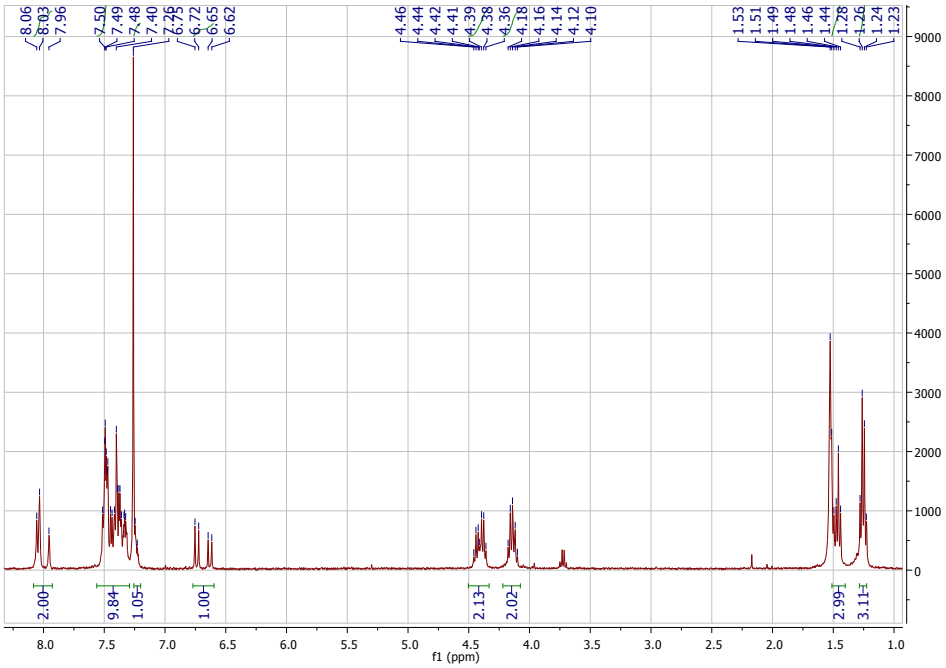


Chemical Formula: $C_{28}H_{24}N_2OS_2$
Molecular Weight: 468.6330

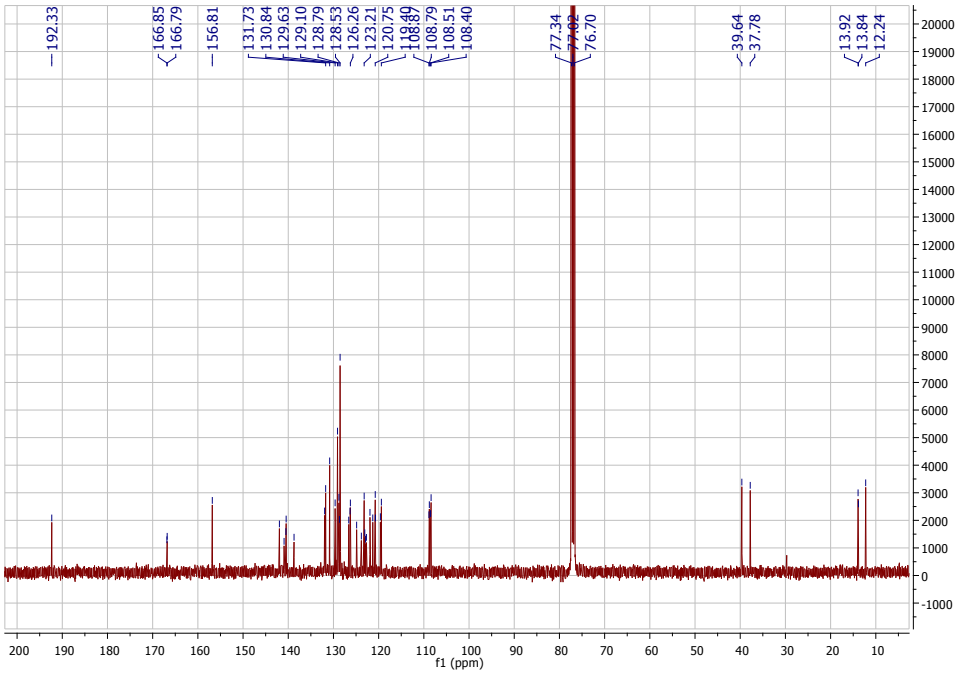


Chemical Formula: $C_{28}H_{24}N_2OS_2$
Molecular Weight: 468.6330

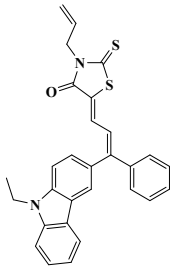
^{1}H NMR spectrum in $CDCl_3$



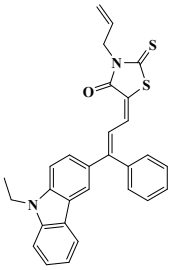
^{13}C NMR spectrum in $CDCl_3$



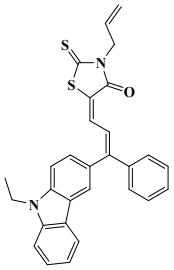
(Z/E)-3-Allyl-5-((Z/E)-3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-2-thioxo-thiazolidin-4-one (L)



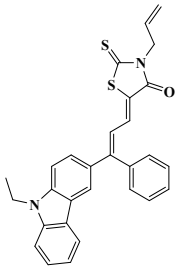
Chemical Formula: $C_{29}H_{24}N_2OS_2$
Molecular Weight: 480.6440



Chemical Formula: $C_{29}H_{24}N_2OS_2$
Molecular Weight: 480.6440

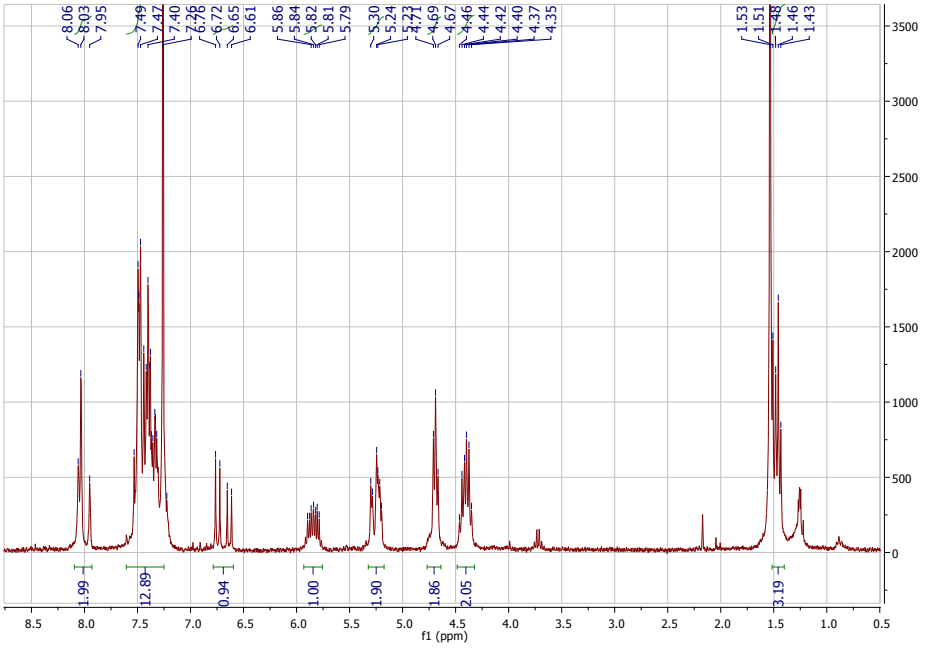


Chemical Formula: $C_{29}H_{24}N_2OS_2$
Molecular Weight: 480.6440

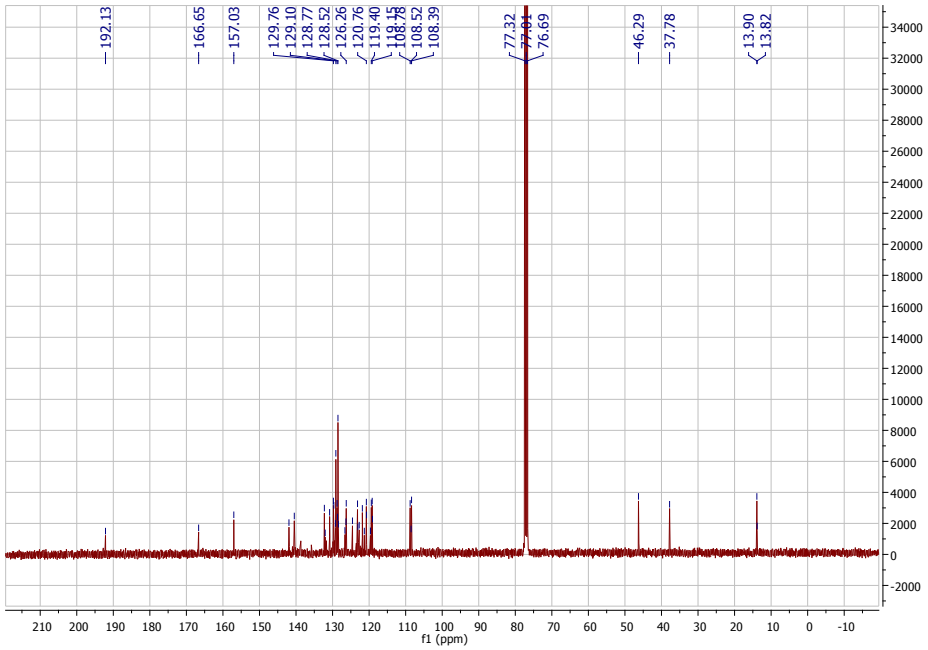


Chemical Formula: $C_{29}H_{24}N_2OS_2$
Molecular Weight: 480.6440

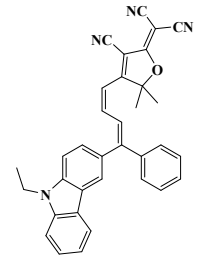
^{1}H NMR spectrum in $CDCl_3$



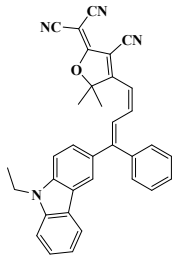
^{13}C NMR spectrum in $CDCl_3$



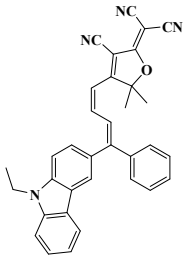
2-(3-Cyano-4-((1Z/*E*,3Z/*E*)-4-(9-ethyl-9*H*-carbazol-3-yl)-4-phenylbuta-1,3-dien-1-yl)-5,5-dimethyl-furan-2(5*H*)-ylidene)malononitrile (M)



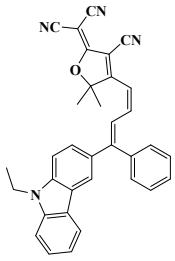
Chemical Formula: $C_{34}H_{26}N_4O$
Molecular Weight: 506.6090



Chemical Formula: $C_{34}H_{26}N_4O$
Molecular Weight: 506.6090

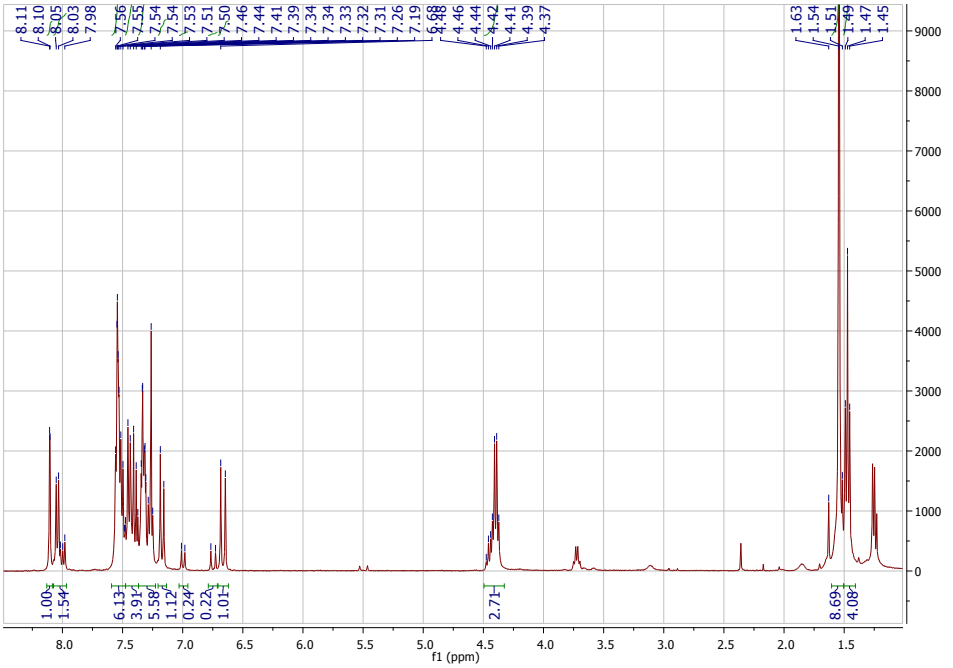


Chemical Formula: $C_{34}H_{26}N_4O$
Molecular Weight: 506.6090

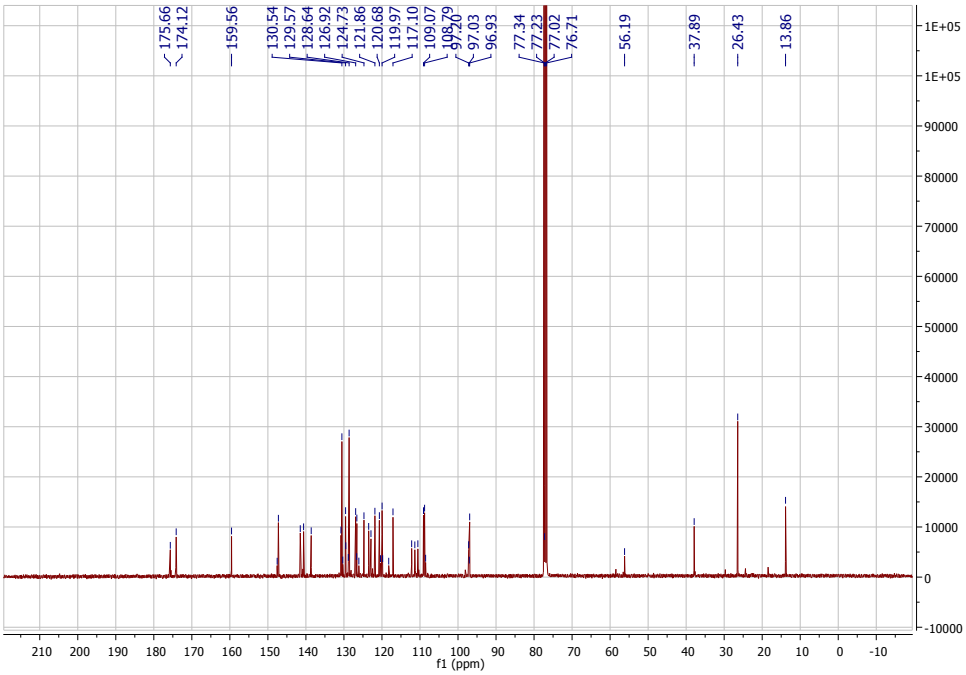


Chemical Formula: $C_{34}H_{26}N_4O$
Molecular Weight: 506.6090

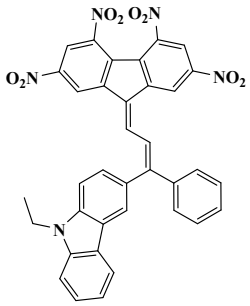
^{1}H NMR spectrum in $CDCl_3$



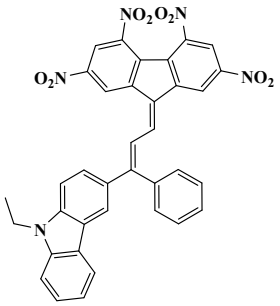
^{13}C NMR spectrum in $CDCl_3$



(Z/E)-9-Ethyl-3-(1-phenyl-3-(2,4,5,7-tetranitro-9*H*-fluoren-9-ylidene)prop-1-en-1-yl)-9*H*-carbazole (N)

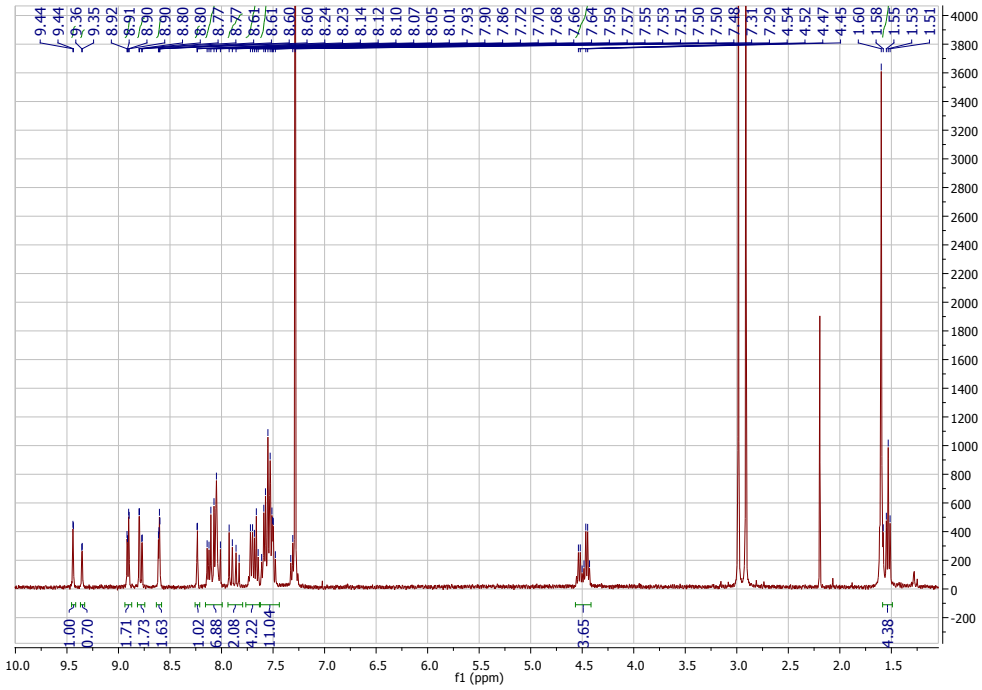


Chemical Formula: $C_{36}H_{23}N_5O_8$
Molecular Weight: 653.6070



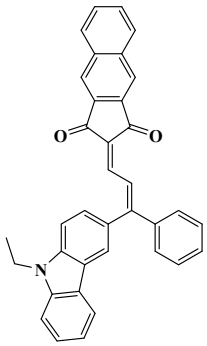
Chemical Formula: $C_{36}H_{23}N_5O_8$
Molecular Weight: 653.6070

^{1}H NMR spectrum in $CDCl_3$

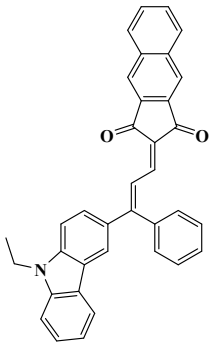


Contour plots of the HOMO and LUMO energy levels of dyes A-N

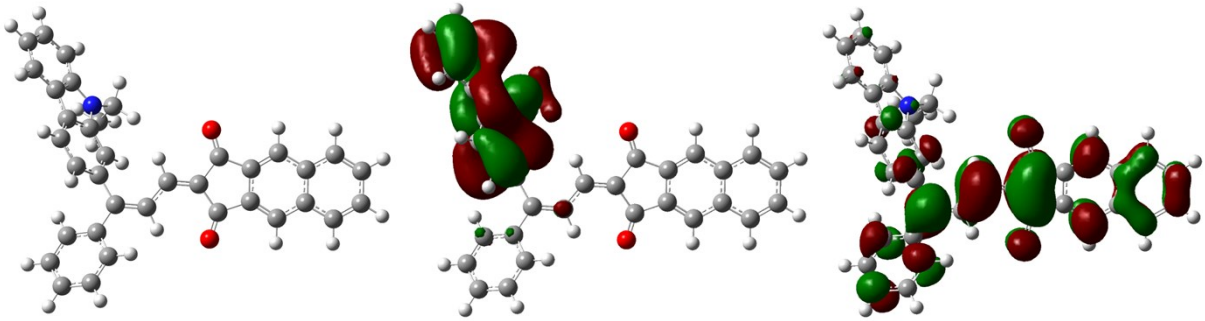
(Z/E)-2-(3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1*H*-cyclopenta[*b*]naphthalene -1,3(2*H*)-dione (A)



Chemical Formula: $C_{36}H_{25}NO_2$
Molecular Weight: 503.6010

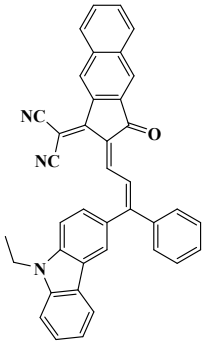


Chemical Formula: $C_{36}H_{25}NO_2$
Molecular Weight: 503.6010

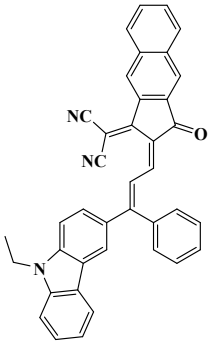


Homo Lumo

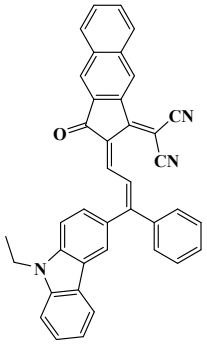
2-((*Z*/*E*)-2-((*Z*/*E*)-3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-3-oxo-2,3-dihydro-1*H*-cyclopenta[*b*]naphthalen-1-ylidene)malononitrile (B)



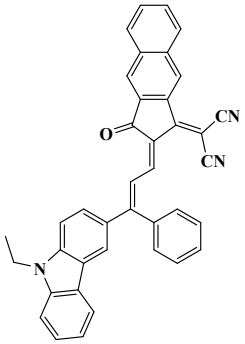
Chemical Formula: $C_{39}H_{25}N_3O$
Molecular Weight: 551.6490



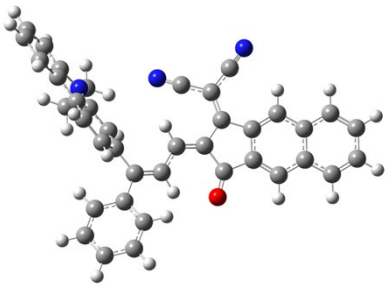
Chemical Formula: $C_{39}H_{25}N_3O$
Molecular Weight: 551.6490

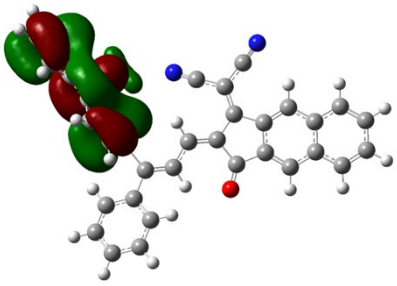


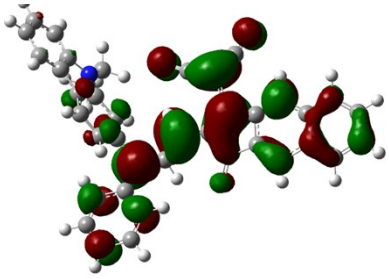
Chemical Formula: $C_{39}H_{25}N_3O$
Molecular Weight: 551.6490



Chemical Formula: $C_{39}H_{25}N_3O$
Molecular Weight: 551.6490

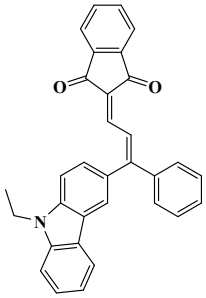




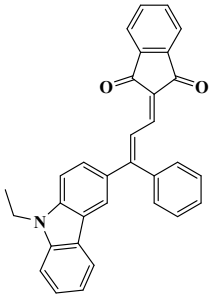


Homo Lumo

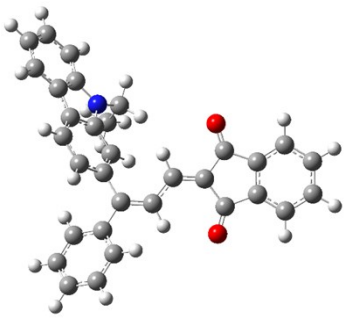
(*Z*/*E*)-2-(3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1*H*-indene-1,3(2*H*)-dione (C/D)

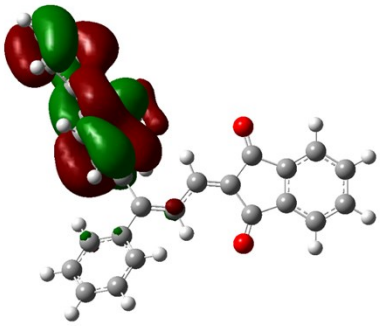


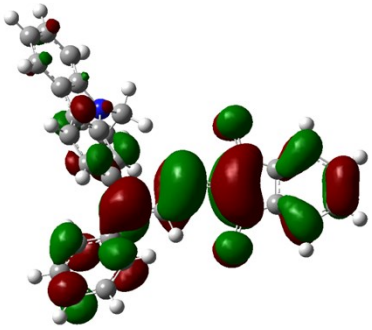
Chemical Formula: $C_{32}H_{23}NO_2$
Molecular Weight: 453.5410



Chemical Formula: $C_{32}H_{23}NO_2$
Molecular Weight: 453.5410

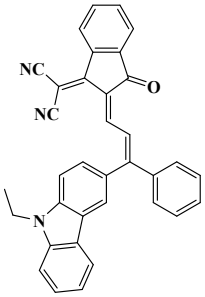




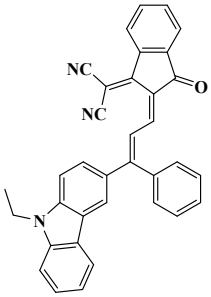


Homo Lumo

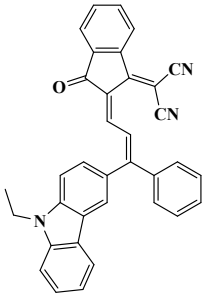
2-((*Z*/*E*)-2-((*Z*/*E*)-3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-3-oxo-2,3-dihydro-1*H*-inden-1-ylidene)malononitrile (E)



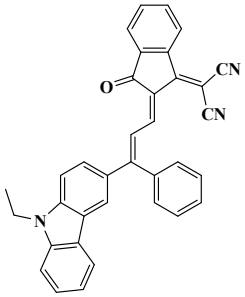
Chemical Formula: $C_{35}H_{23}N_3O$
Molecular Weight: 501.5890



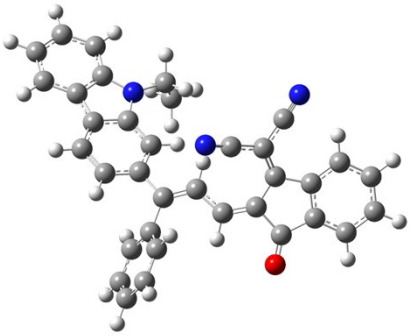
Chemical Formula: $C_{35}H_{23}N_3O$
Molecular Weight: 501.5890

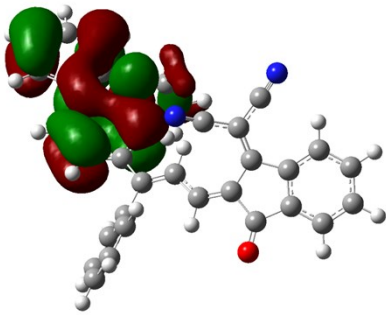


Chemical Formula: $C_{35}H_{23}N_3O$
Molecular Weight: 501.5890

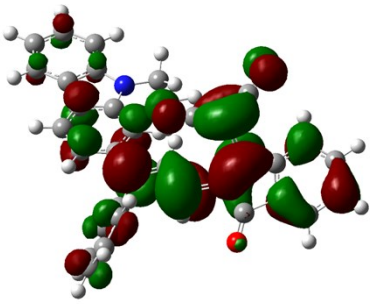


Chemical Formula: $C_{35}H_{23}N_3O$
Molecular Weight: 501.5890



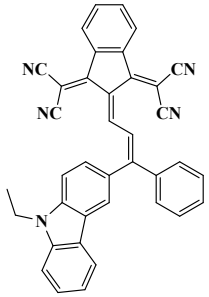


Homo

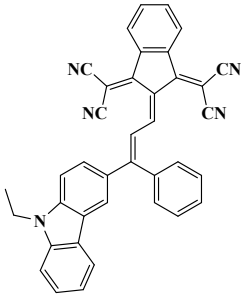


Lumo

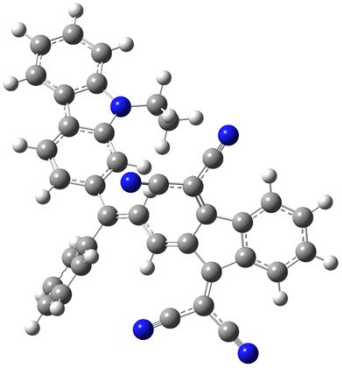
(*Z*/*E*)-2,2'-(2-(3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1*H*-indene-1,3(2*H*)-diylidene)dimalononitrile (F)

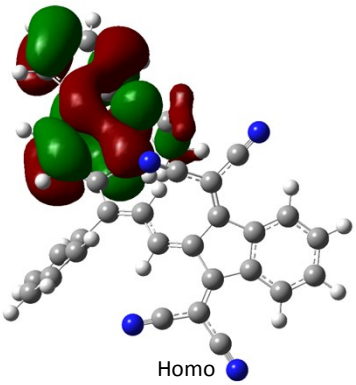


Chemical Formula: $C_{38}H_{23}N_5$
Molecular Weight: 549.6370

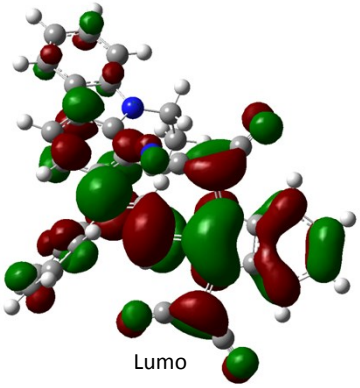


Chemical Formula: $C_{38}H_{23}N_5$
Molecular Weight: 549.6370



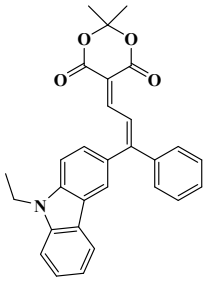


Homo

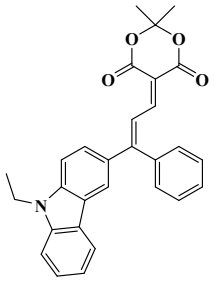


Lumo

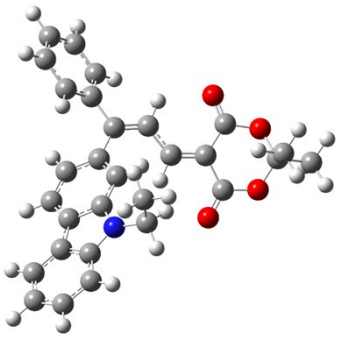
(Z/E)-5-(3-(9-Ethyl-9H-carbazol-3-yl)-3-phenylallylidene)-2,2-dimethyl-1,3-dioxane-4,6-dione (G)

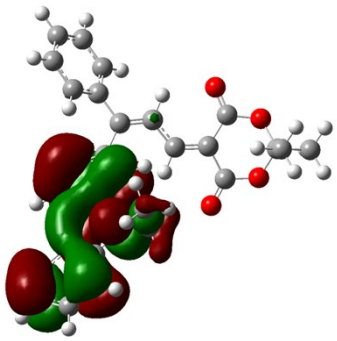


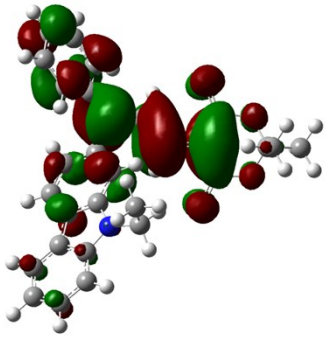
Chemical Formula: $C_{29}H_{25}NO_4$
Molecular Weight: 451.5220



Chemical Formula: $C_{29}H_{25}NO_4$
Molecular Weight: 451.5220

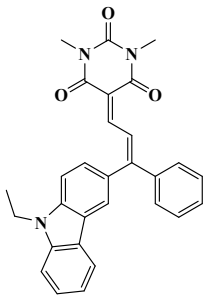




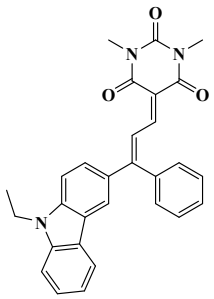


Homo Lumo

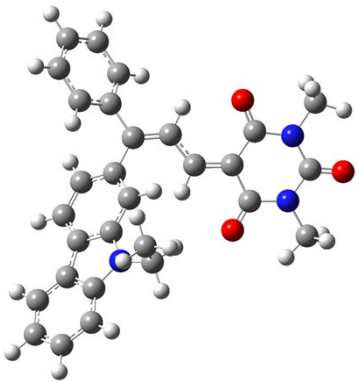
(*Z*/*E*)-5-(3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1,3-dimethylpyrimidine-2,4,6(1*H*,3*H*,5*H*)-trione (H/I)

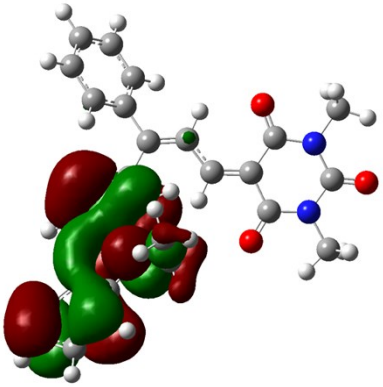


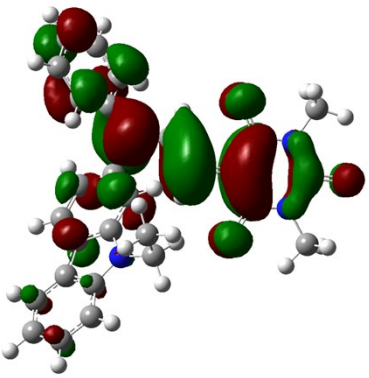
Chemical Formula: $C_{29}H_{25}N_3O_3$
Molecular Weight: 463.5370



Chemical Formula: $C_{29}H_{25}N_3O_3$
Molecular Weight: 463.5370

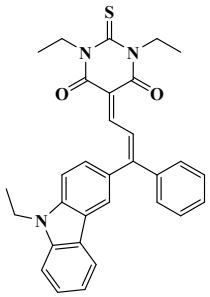




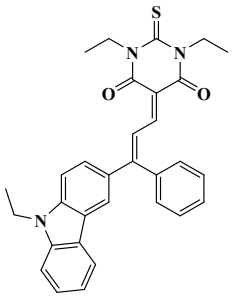


Homo Lumo

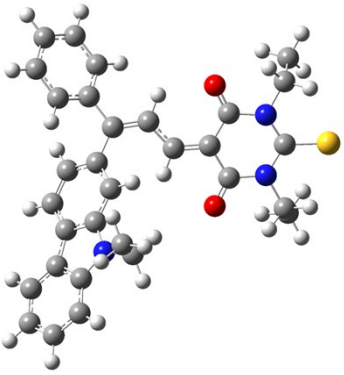
(*Z*/*E*)-1,3-Diethyl-5-(3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-2-thioxodihydro-pyrimidine-4,6(1*H*,5*H*)-dione (J)

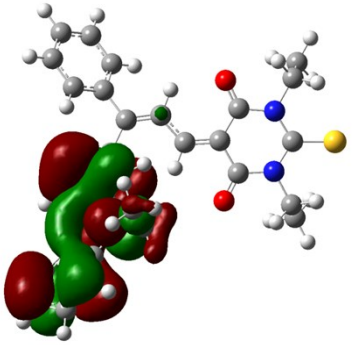


Chemical Formula: $C_{31}H_{29}N_3O_2S$
Molecular Weight: 507.6520

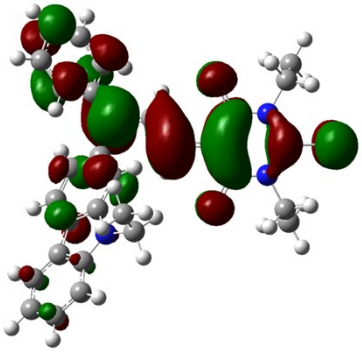


Chemical Formula: $C_{31}H_{29}N_3O_2S$
Molecular Weight: 507.6520



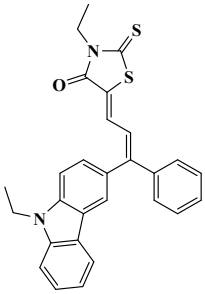


Homo

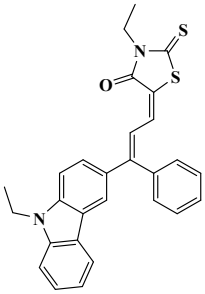


Lumo

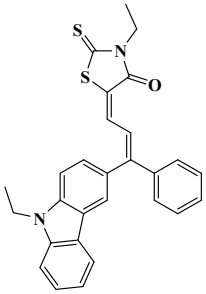
(Z/E)-3-Ethyl-5-((Z/E)-3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-2-thioxo-thiazolidin-4-one (K)



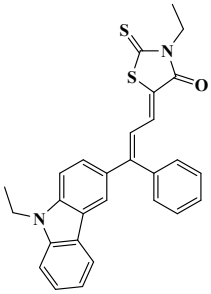
Chemical Formula: $C_{28}H_{24}N_2OS_2$
Molecular Weight: 468.6330



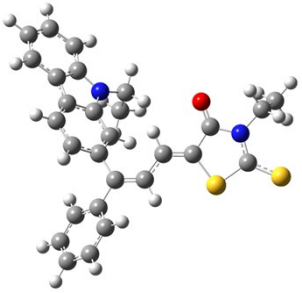
Chemical Formula: $C_{28}H_{24}N_2OS_2$
Molecular Weight: 468.6330

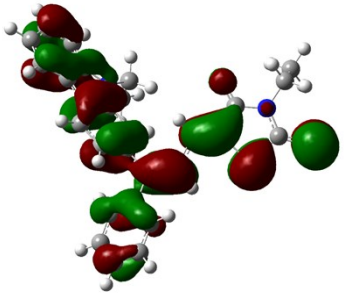


Chemical Formula: $C_{28}H_{24}N_2OS_2$
Molecular Weight: 468.6330

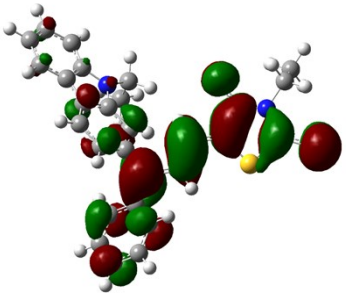


Chemical Formula: $C_{28}H_{24}N_2OS_2$
Molecular Weight: 468.6330



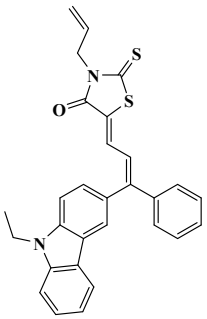


Homo

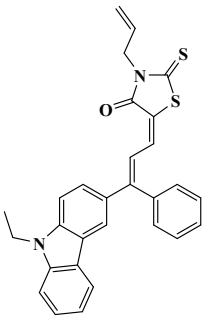


Lumo

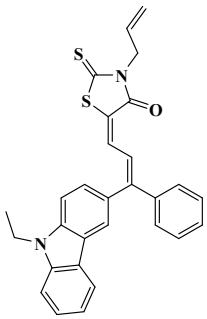
(Z/E)-3-Allyl-5-((Z/E)-3-(9-ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-2-thioxo-thiazolidin-4-one (L)



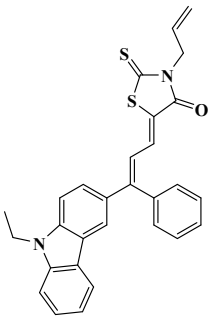
Chemical Formula: $C_{29}H_{24}N_2OS_2$
Molecular Weight: 480.6440



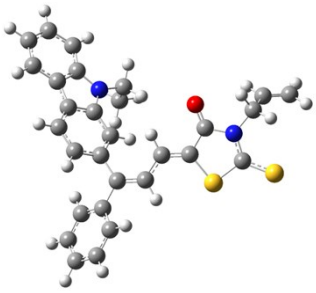
Chemical Formula: $C_{29}H_{24}N_2OS_2$
Molecular Weight: 480.6440

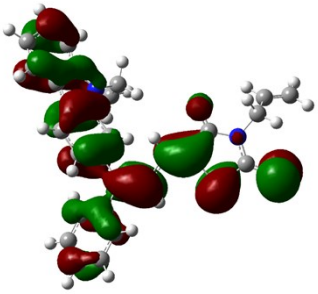


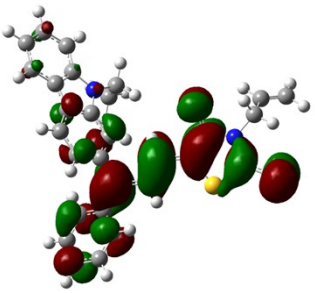
Chemical Formula: $C_{29}H_{24}N_2OS_2$
Molecular Weight: 480.6440



Chemical Formula: $C_{29}H_{24}N_2OS_2$
Molecular Weight: 480.6440

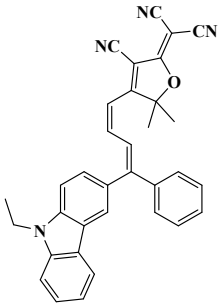




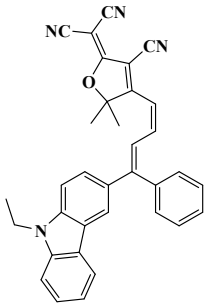


Homo Lumo

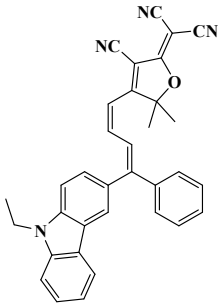
2-(3-Cyano-4-((1Z/*E*,3Z/*E*)-4-(9-ethyl-9*H*-carbazol-3-yl)-4-phenylbuta-1,3-dien-1-yl)-5,5-dimethyl-furan-2(5*H*)-ylidene)malononitrile (M)



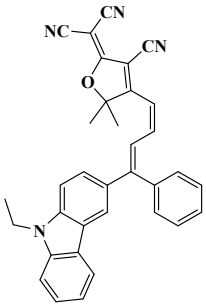
Chemical Formula: $C_{34}H_{26}N_4O$
Molecular Weight: 506.6090



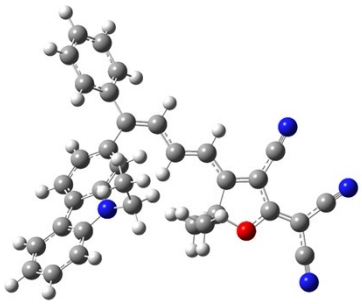
Chemical Formula: $C_{34}H_{26}N_4O$
Molecular Weight: 506.6090

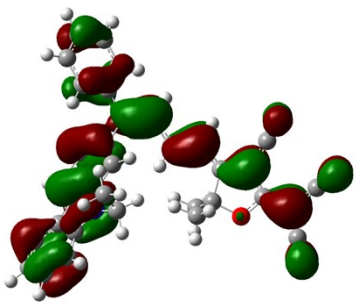


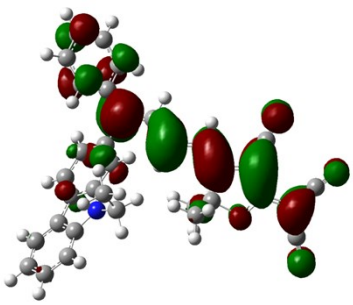
Chemical Formula: $C_{34}H_{26}N_4O$
Molecular Weight: 506.6090



Chemical Formula: $C_{34}H_{26}N_4O$
Molecular Weight: 506.6090

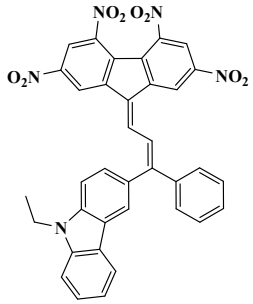




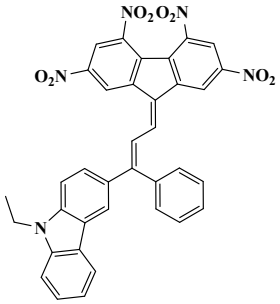


Homo Lumo

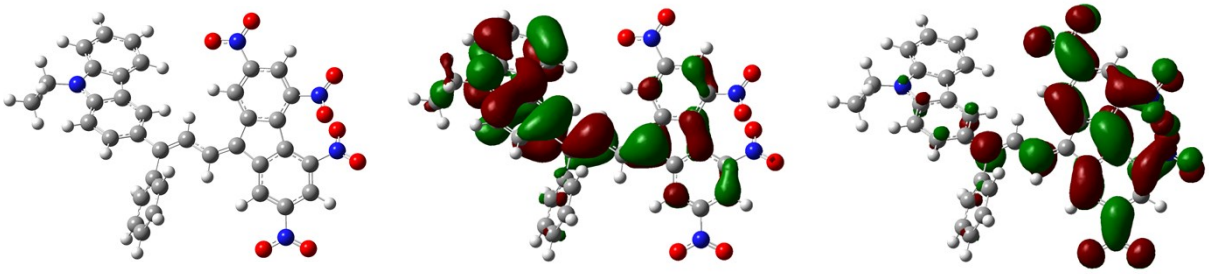
(Z/E)-9-Ethyl-3-(1-phenyl-3-(2,4,5,7-tetranitro-9*H*-fluoren-9-ylidene)prop-1-en-1-yl)-9*H*-carbazole (N)



Chemical Formula: $C_{36}H_{23}N_5O_8$
Molecular Weight: 653.6070



Chemical Formula: $C_{36}H_{23}N_5O_8$
Molecular Weight: 653.6070



Homo Lumo

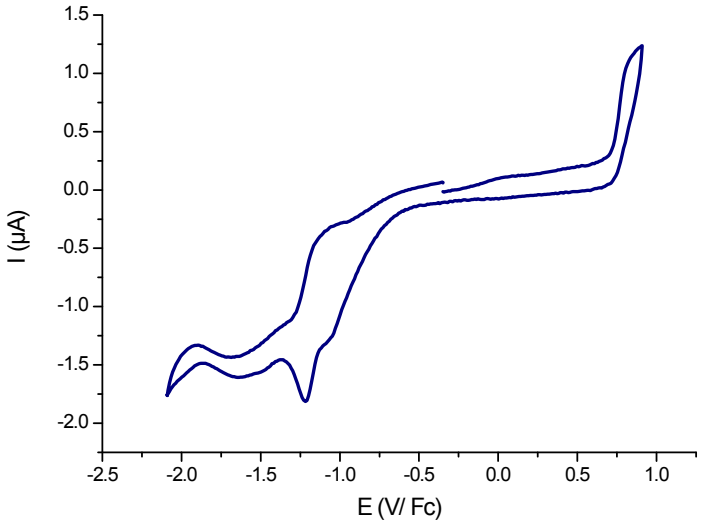
Energy levels of the main orbitals for dyes A-N

	Homo-1	Homo	Lumo	Lumo+1
C/D	-5,690	-5,563	-2,636	-1,964
E	-5,781	-5,715	-3,061	-2,491
F	-5,901	-5,834	-3,275	-3,151
N	-6,413	-5,898	-3,911	-3,293
H/I	-5,818	-5,637	-2,792	-1,048
J	-5,770	-5,706	-2,996	-1,245
M	-6,023	-5,949	-3,332	-1,600
G	-5,896	-5,686	-2,784	-1,058
A	-5,671	-5,553	-2,725	-1,780
B	-5,714	-5,479	-3,172	-2,428
K	-5,683	-5,664	-2,719	-1,361
L	-5,685	-5,667	-2,725	-1,364

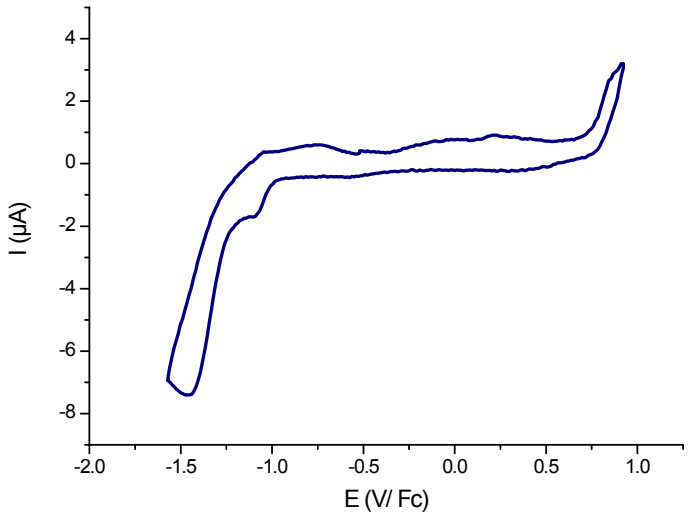
Cyclic voltammograms of the push pull compounds

The electrochemical properties of the investigated compounds were measured in dichloromethane by cyclic voltammetry, scan rate 100 $mV.s^{-1}$, with tetrabutylammonium perchlorate (0.1M) as a supporting electrolyte in a standard one-compartment, three-electrode electrochemical cell under an argon stream using a VSP BioLogic potentiostat. The working, pseudo-reference and counter electrodes were platinum disk (Ø = 1 mm), Ag wire, and Au wire gauze, respectively. Ferrocene/ferrocenium was used as the internal standard.

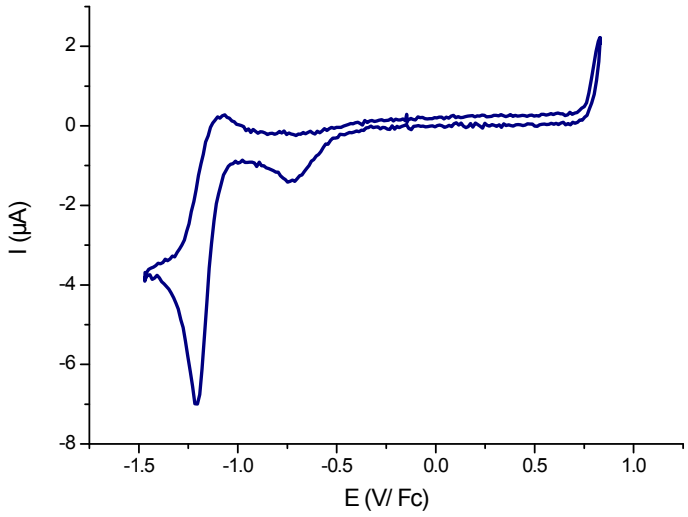
Compound A:



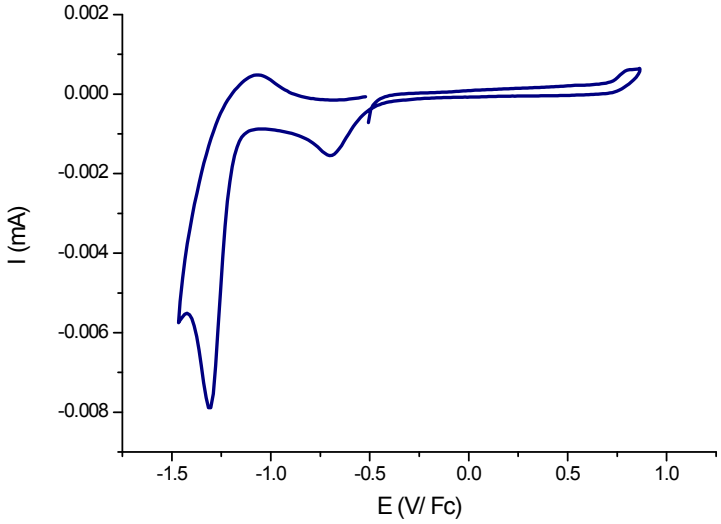
Compound B :



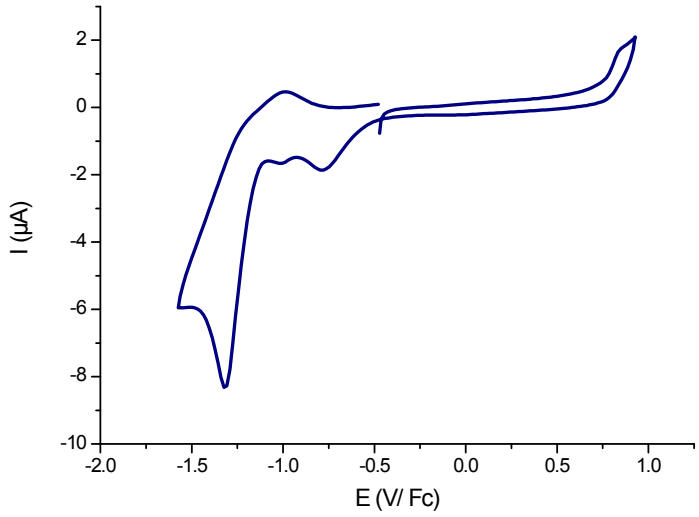
Compound C :



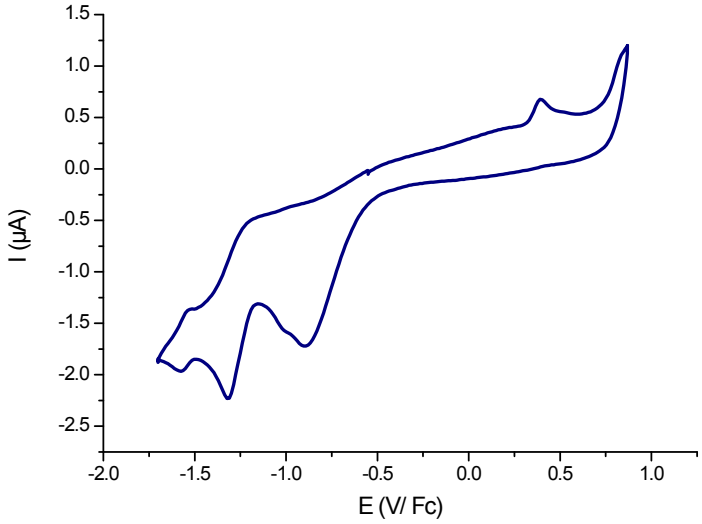
Compound D :



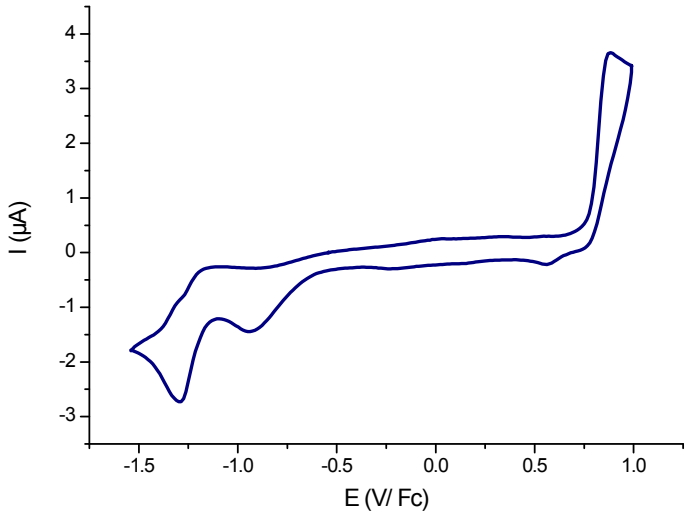
Compound E :



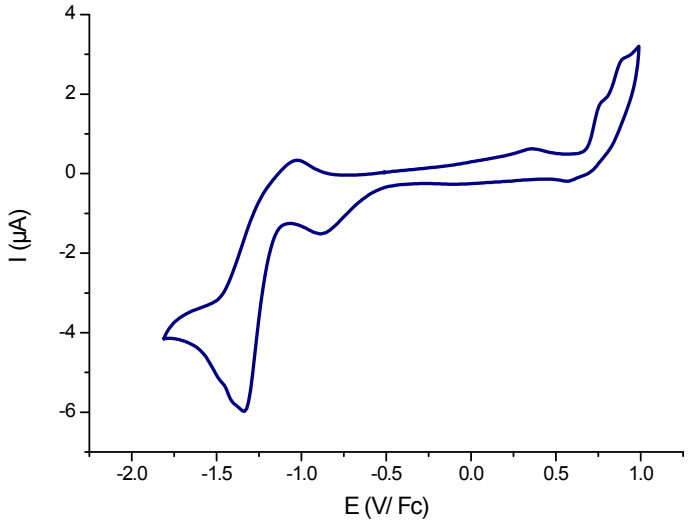
Compound F :



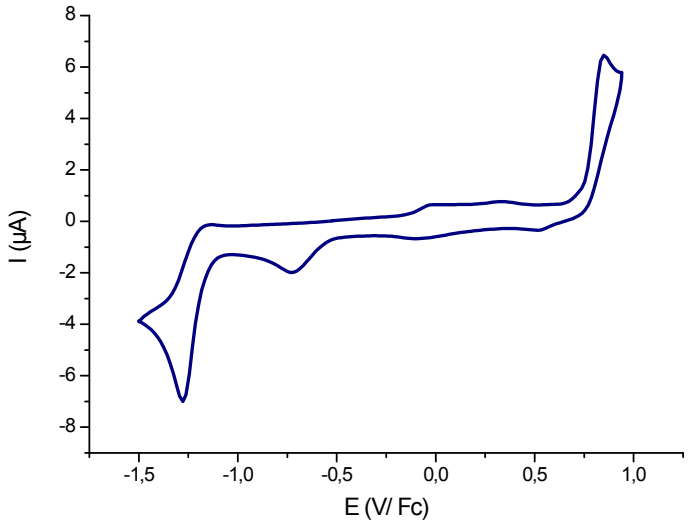
Compound G :



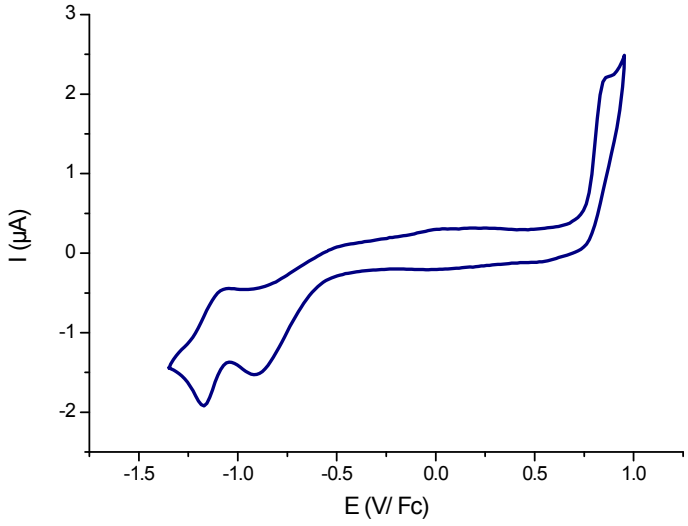
Compound H :



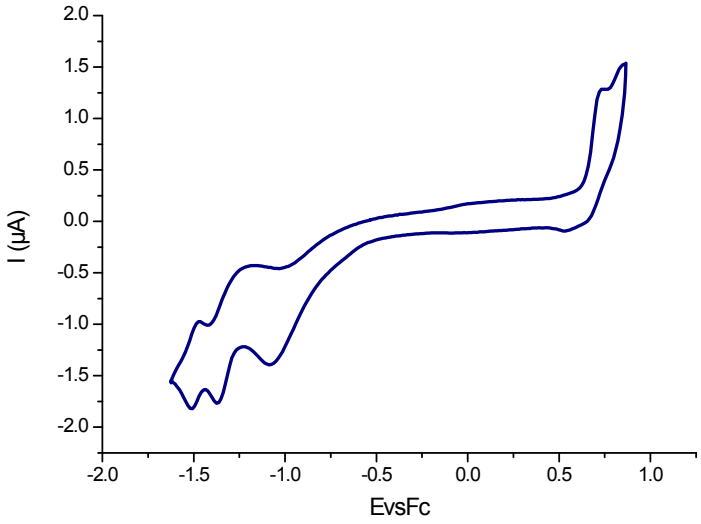
Compound I :



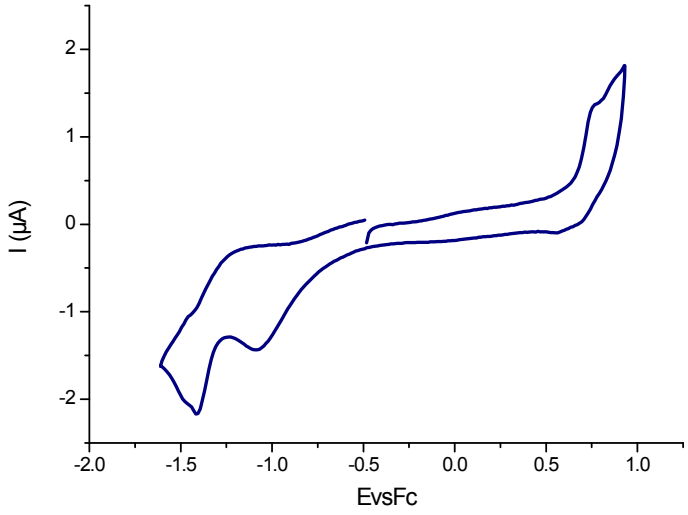
Compound J :



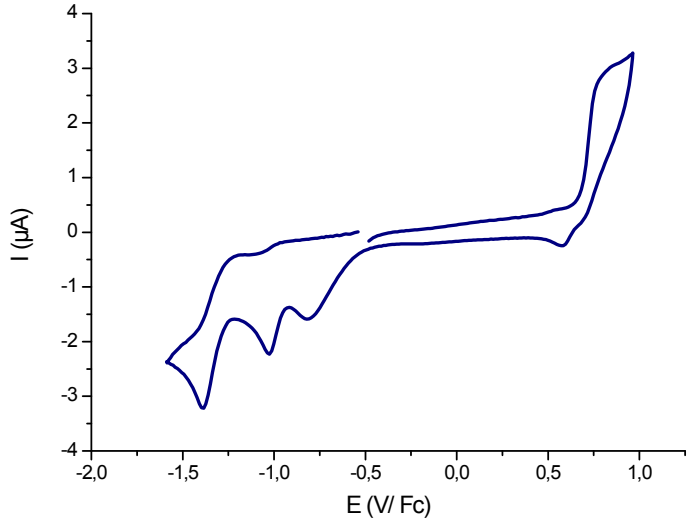
Compound K :



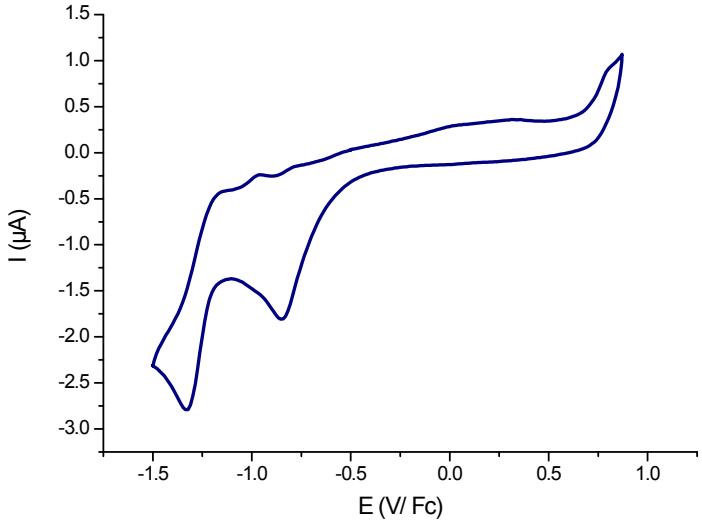
Compound L :



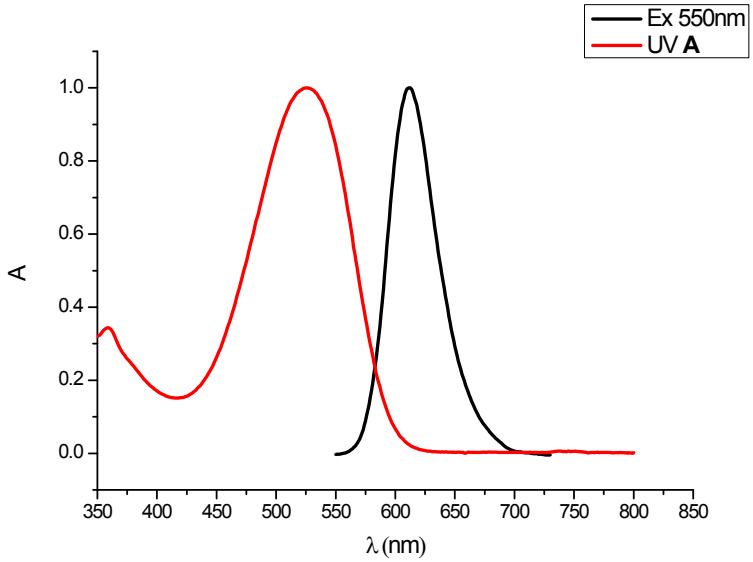
Compound M :

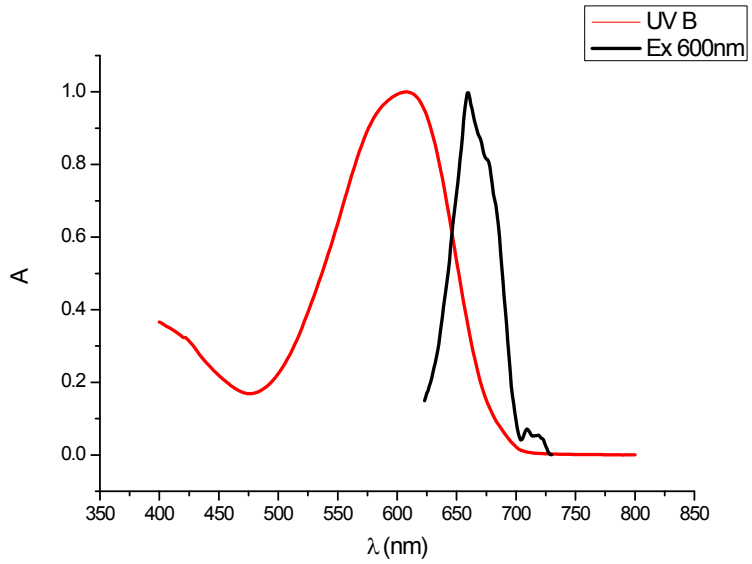


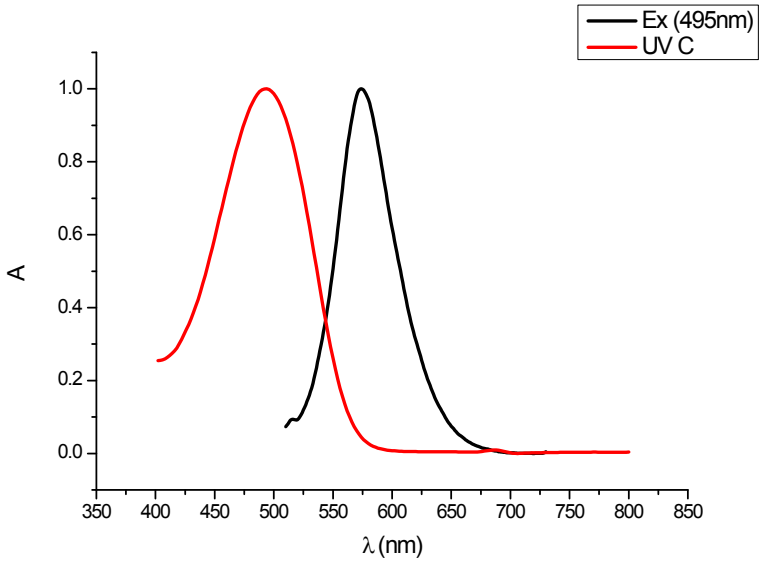
Compound N :

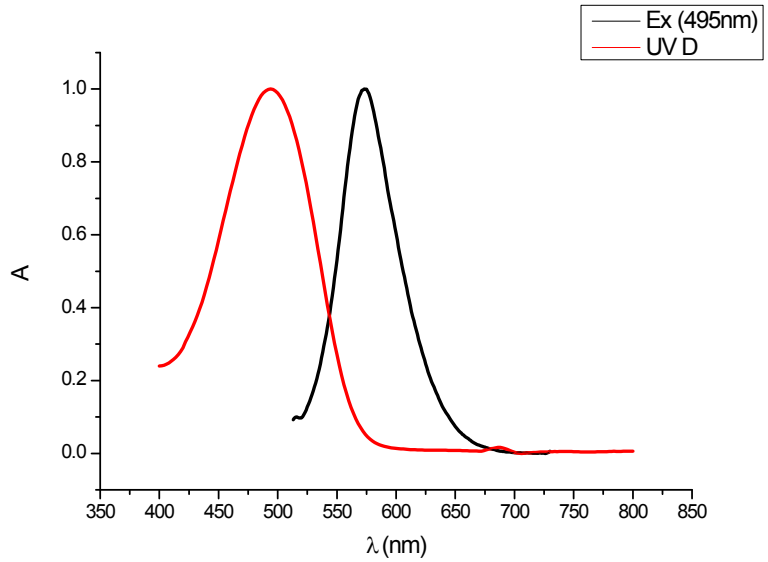


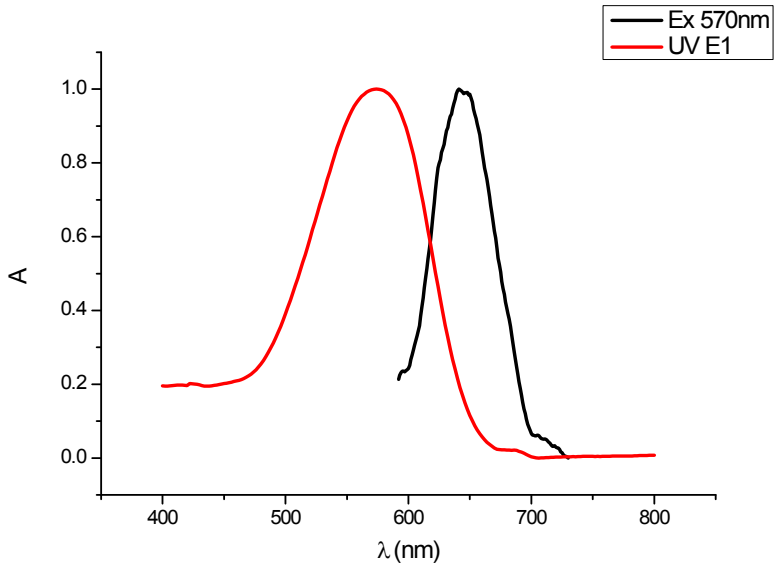
Fluorescence spectra of F, G, I, J, K and M in chloroform recorded upon photoexcitation at different wavelengths in chloroform.

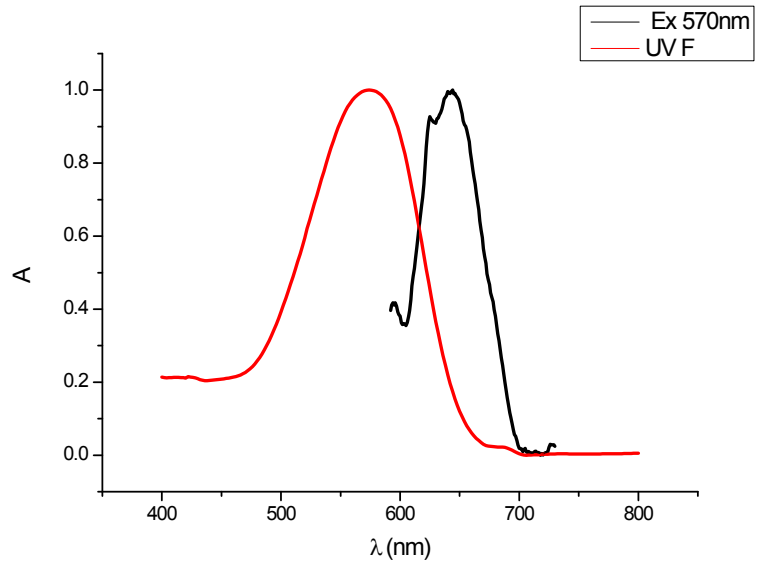


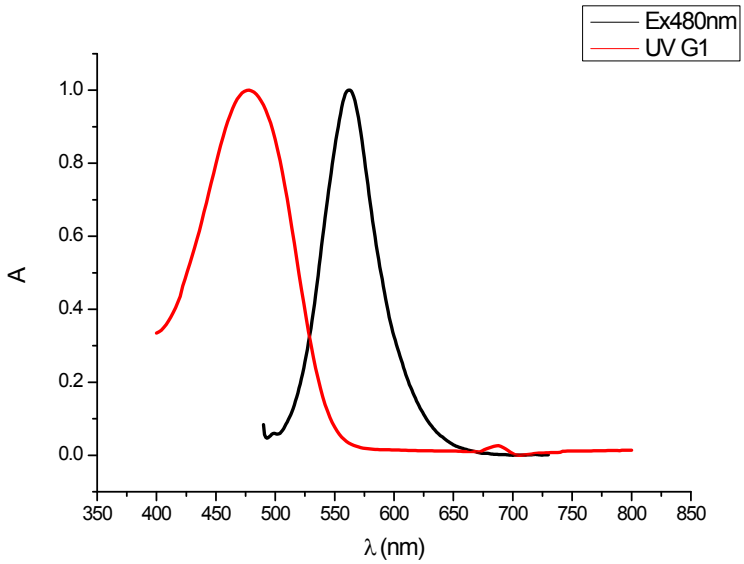


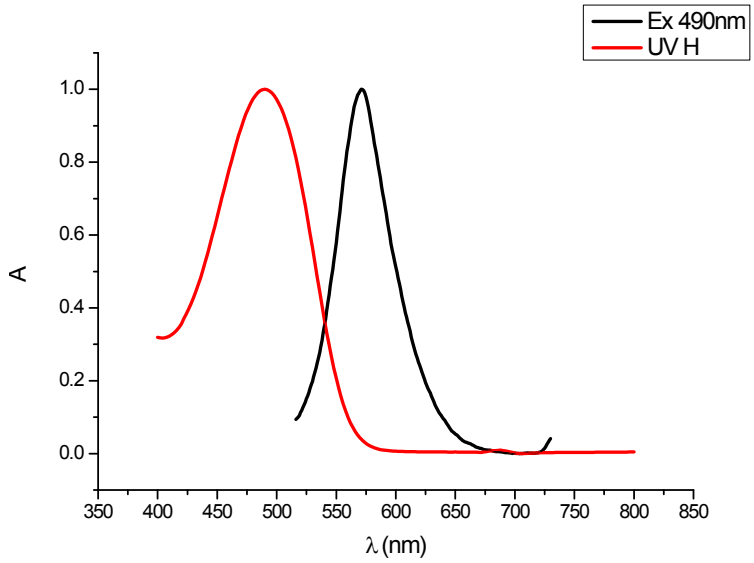


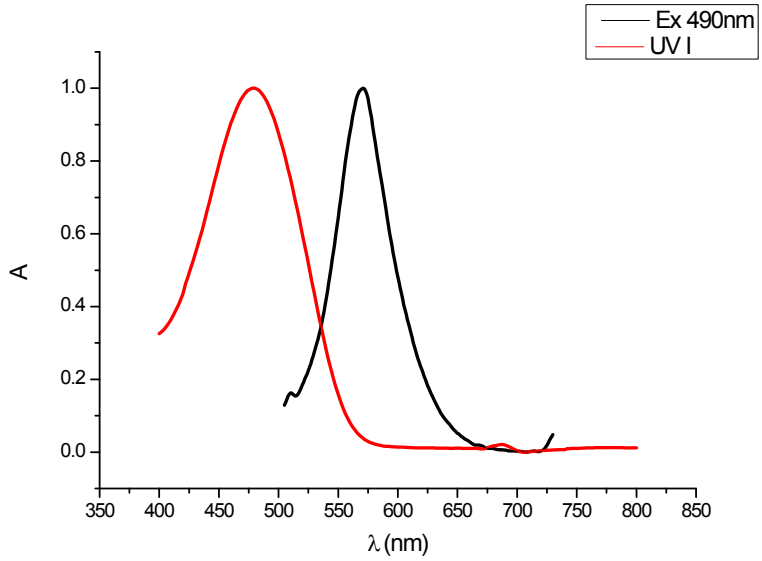

Ex (495nm)
UV D
A
λ (nm)

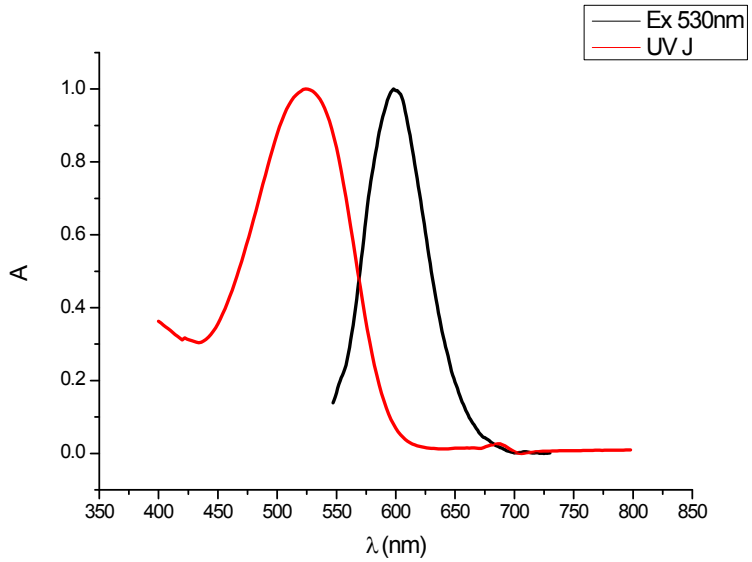

Ex 570nm
UV E1
A
λ (nm)

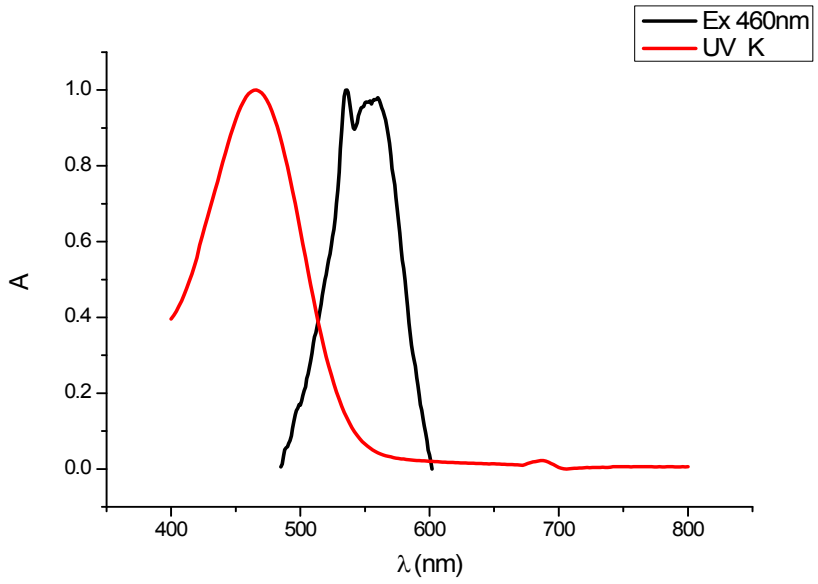

Ex 570nm
UV F
A
λ (nm)

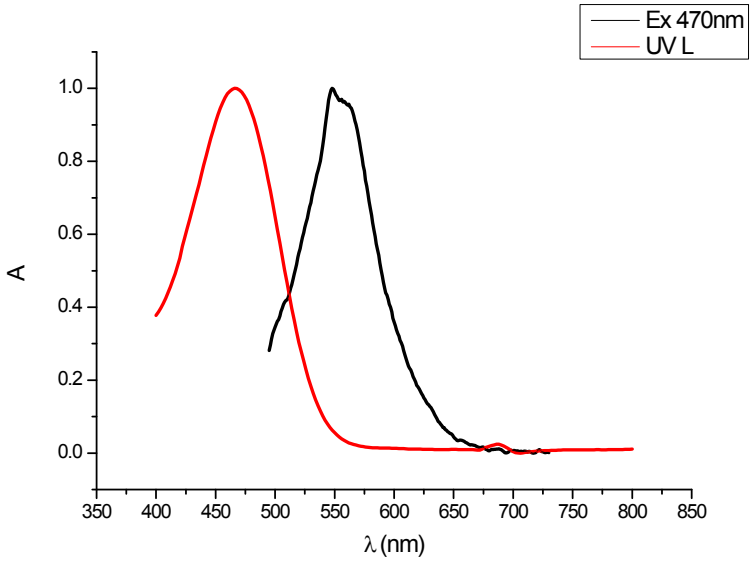

Ex480nm
UV G1
A
λ (nm)

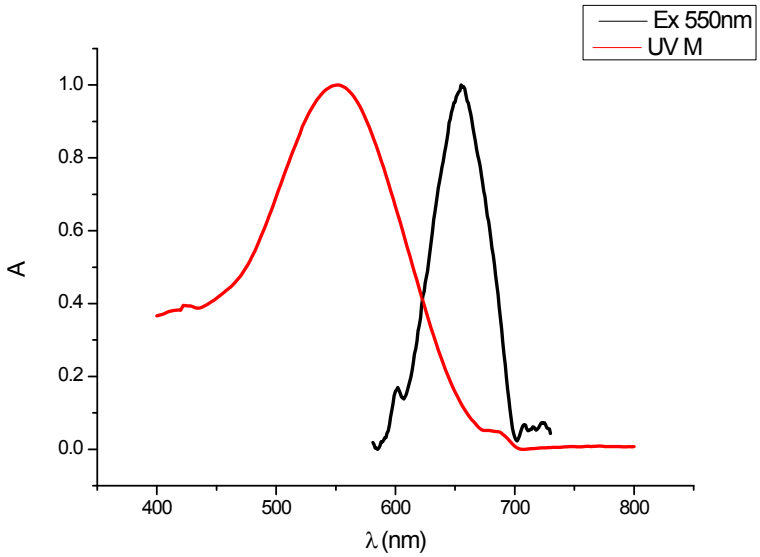

Ex 490nm
UV H
A
λ (nm)


Ex 490nm
UV I
A
λ (nm)


Ex 530nm
UV J
A
λ (nm)

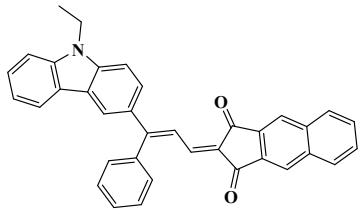

Ex 460nm
UV K
A
λ (nm)


Ex 470nm
UV L
A
λ (nm)

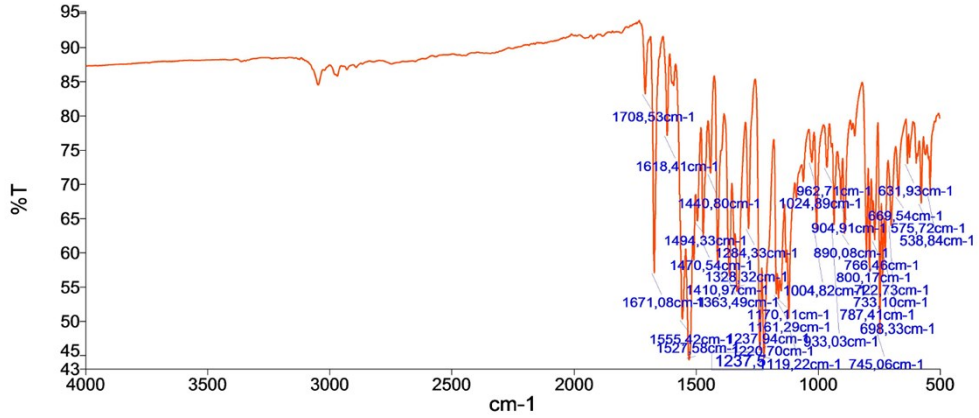

Ex 550nm
UV M
1.0
0.8
0.6
0.4
0.2
0.0
A
400
500
600
700
800
λ (nm)

IR spectra of all compounds, obtained which an ATR PerkinElmer instrument

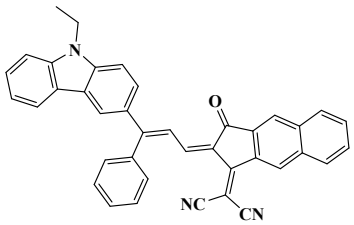
Compound A



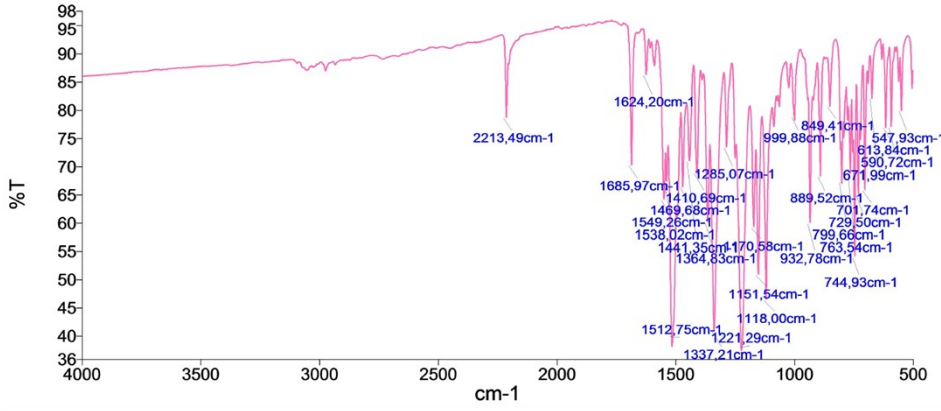
Chemical Formula: $C_{36}H_{25}NO_2$
Molecular Weight: 503.6010



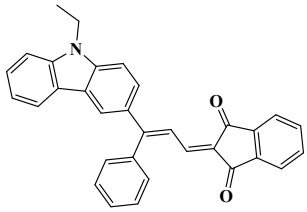
Compound B



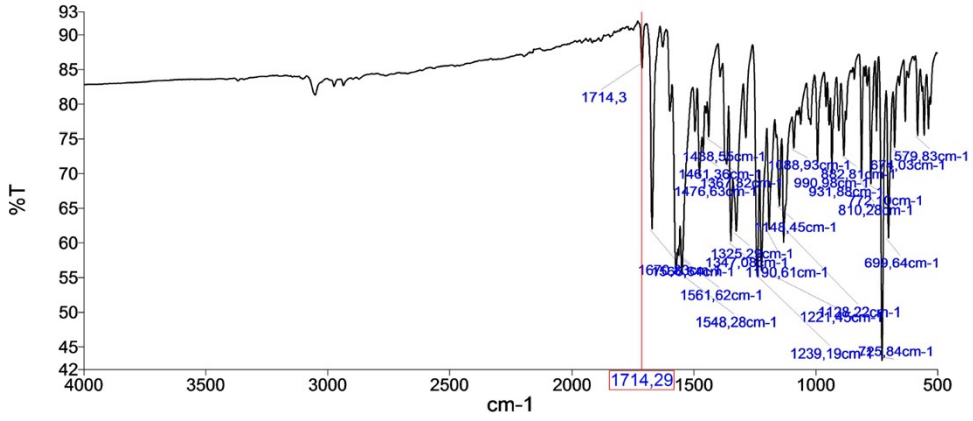
Chemical Formula: $C_{39}H_{25}N_3O$
Molecular Weight: 551.6490



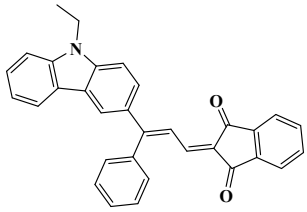
Compound C



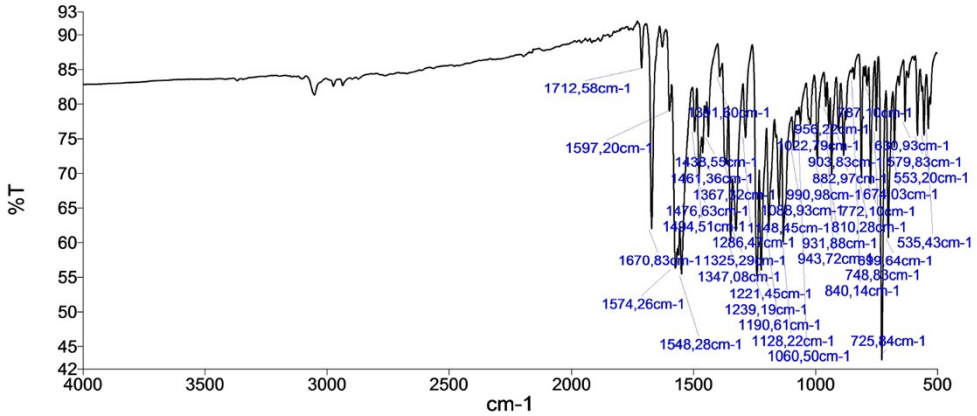
Chemical Formula: $C_{32}H_{23}NO_2$
Molecular Weight: 453.5410



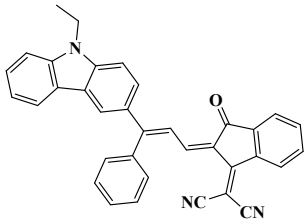
Compound D



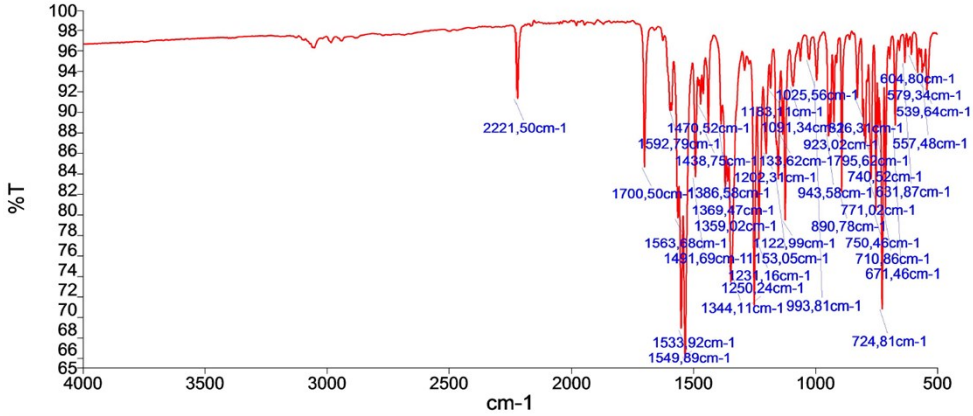
Chemical Formula: $C_{32}H_{23}NO_2$
Molecular Weight: 453.5410



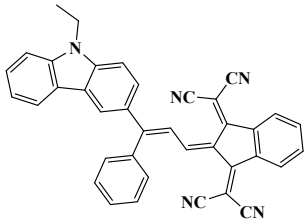
Compound E



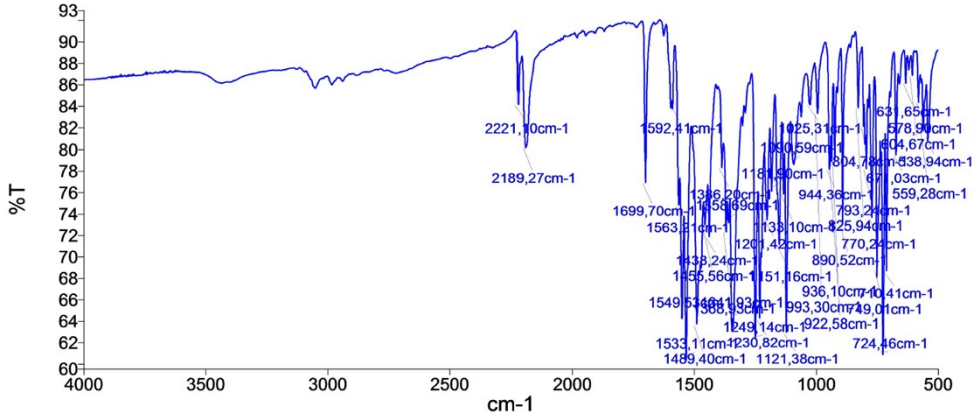
Chemical Formula: $C_{35}H_{23}N_3O$
Molecular Weight: 501.5890



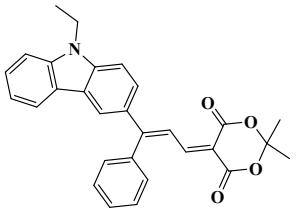
Compound F



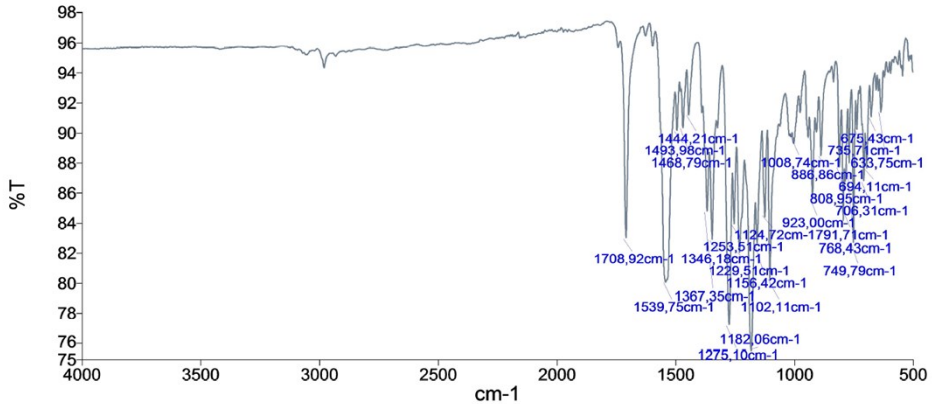
Chemical Formula: $C_{38}H_{23}N_5$
Molecular Weight: 549.6370



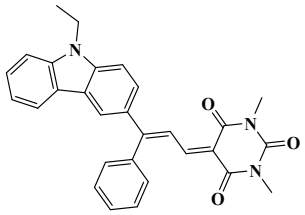
Compound G



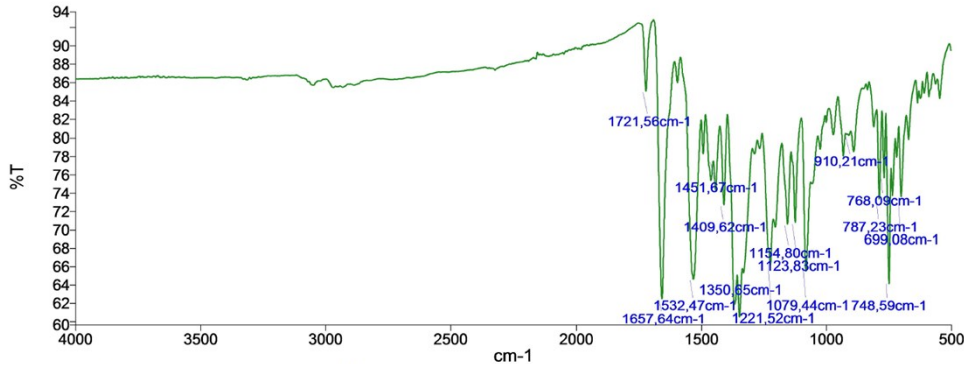
Chemical Formula: $C_{29}H_{25}NO_4$
Molecular Weight: 451.5220



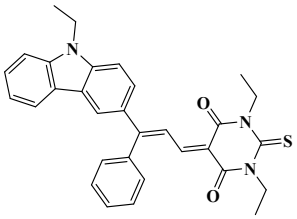
Compound H (mixture of isomers)



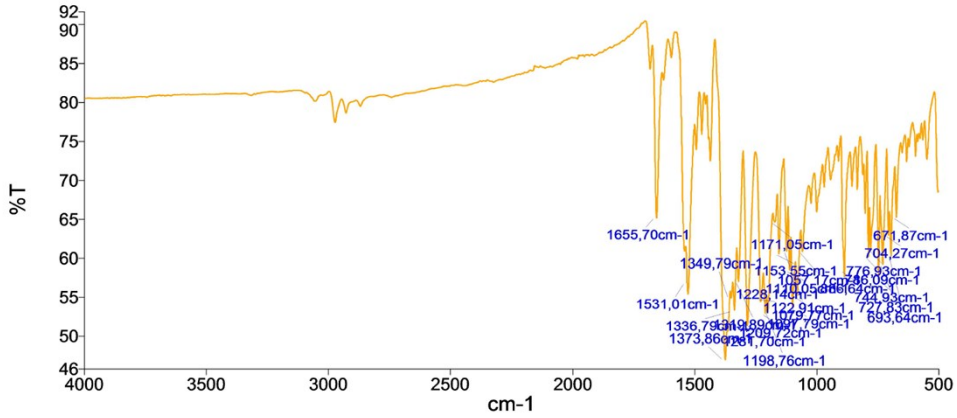
Chemical Formula: $C_{29}H_{25}N_3O_3$
Molecular Weight: 463.5370



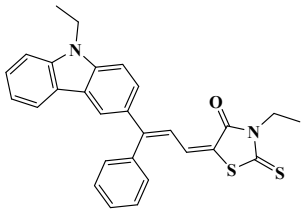
Compound J



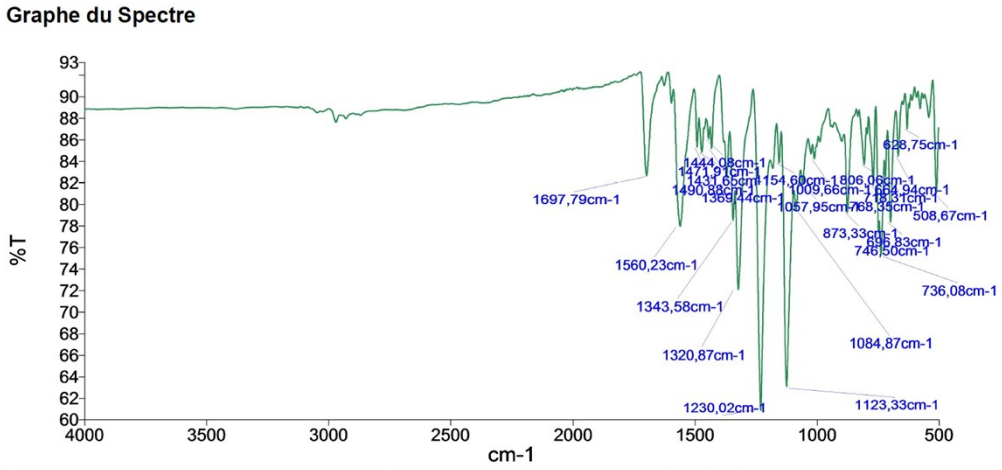
Chemical Formula: $C_{31}H_{29}N_3O_2S$
Molecular Weight: 507.6520



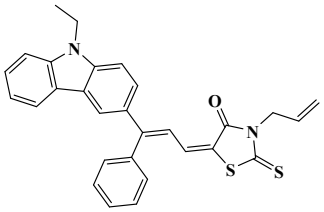
Compound K



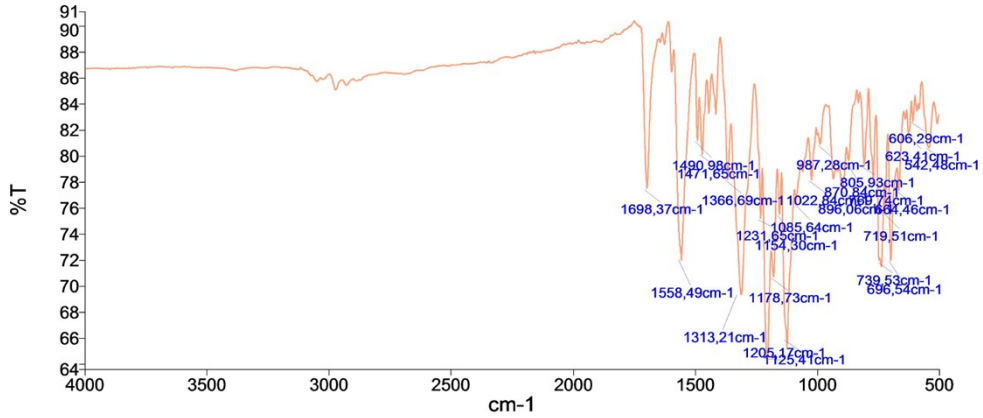
Chemical Formula: $C_{28}H_{24}N_2OS_2$
Molecular Weight: 468.6330



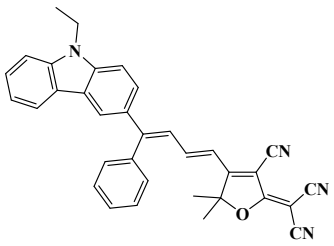
Compound L



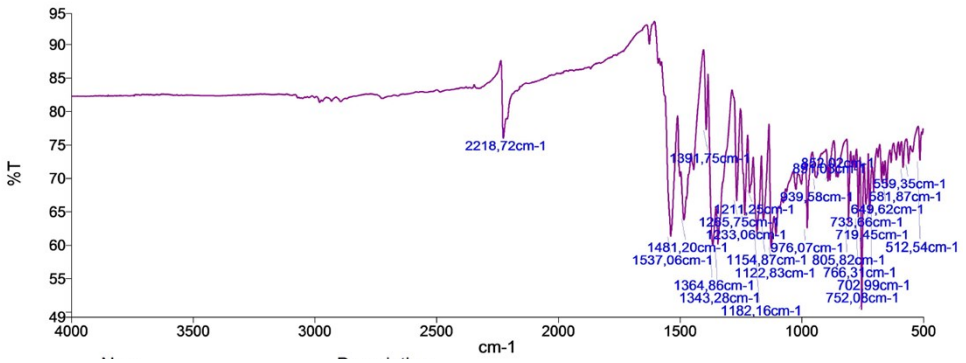
Chemical Formula: $C_{29}H_{24}N_2OS_2$
Molecular Weight: 480.6440



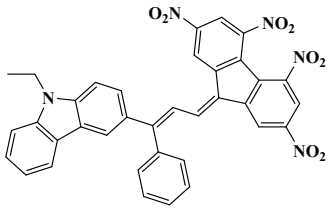
Compound M



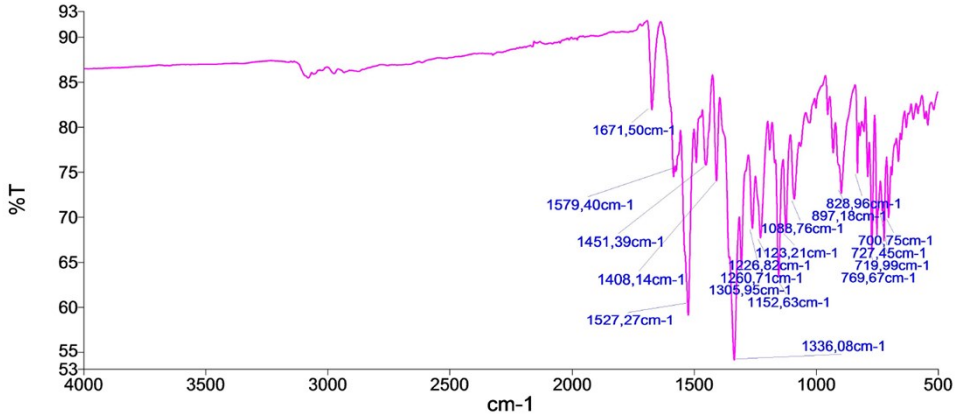
Chemical Formula: $C_{34}H_{26}N_4O$
Molecular Weight: 506.6090



Compound N

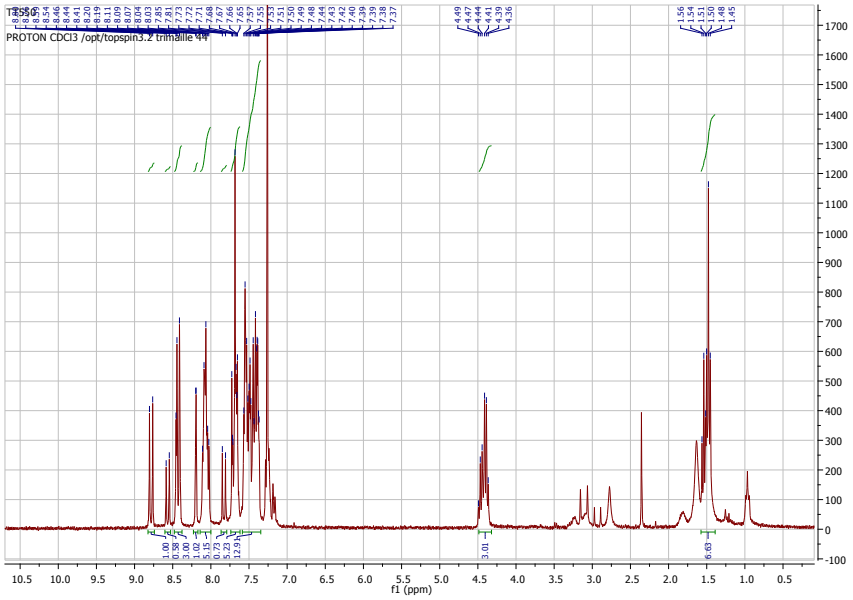


Chemical Formula: $C_{36}H_{23}N_5O_8$
Molecular Weight: 653.6070



Iodine-catalyzed thermal cis/trans isomerization experiments in toluene.

^{1}H NMR spectrum in $CDCl_3$ of (Z/E)-2-(3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1*H*-cyclopenta[*b*]naphthalene -1,3(2*H*)-dione (A) after a tentative of thermal isomerization with iodine in toluene upon overnight reflux. (SI 3)



^{1}H NMR spectrum in $CDCl_3$ of (Z/E)-2-(3-(9-Ethyl-9*H*-carbazol-3-yl)-3-phenylallylidene)-1*H*-cyclopenta[*b*]naphthalene -1,3(2*H*)-dione (A) before iodine-catalyzed thermal isomerization (SI 3)