

Full Length Research Paper

Solvent effects and photophysical studies of ADS560EI laser dye

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The ground state (μ_g) and the excited state (μ_e) dipole moments of a laser dye ADS560EI is estimated in various solvents by Lippert, Backshieve, Kawski-Chamma-Viallet, McRae and Suppan equations using the variation of stokes shift with the solvent dielectric constant and refractive index. The excited state dipole moments were also calculated by using the variation of stokes shift with microscopic solvent polarity parameter (E_t^N). It was observed that the dipole moment values of excited state (μ_e) were higher than corresponding ground state values (μ_g), indicating a substantial redistribution of the π -electron densities in a more polar excited state than the ground state. The rotational reorientation of this probe molecule has also been examined in various solvents. It is observed that the increase in viscosity of the solvent is responsible for the increase in the rotational relaxation time of the probe molecule.

Key words: Stokes shift, excited state, ground state, dipole moment, reorientation time.

INTRODUCTION

Molecular orientation in a liquid is greatly influenced by its environment, as molecules are continuously interacting with each other due to their close proximity. By understanding the molecular rotation in liquids, one can get a better insight into the nature of the solute-solvent interactions, which in turn creates a lot of interest in understanding the photo physical properties of molecules.

A molecule rotating in a liquid continuously experiences friction, due to its volume, size, viscosity, etc. The general approach is to measure the reorientation time of a solute molecule in a liquid experimentally and model its rotation using diffusion based theories like Stokes Einstein Debye (SED)(1929); Nee-Zwanzig (1970); Van der Zwan and Hynes (1985); Alavi-Waldeck (1993). The two models Nee-Zwanzig and Van der Zwan-Hynes, which treat the solute as a point dipole, could not explain the observed trend qualitatively. But Alavi-Waldeck model which treats the solutes electrostatic field as a distribution of charges is able to explain the observed behavior qualitatively.

According to SED theory, the reorientation time of solute molecule is proportional to its size, volume, viscosity and surrounding temperature. However, this reorientation of a molecule can be better understood if we analyze the molecule under solvent effects. Various techniques are available; among the most popular is the Lippert's, Bakhshiev's, Kawski-Chamma-Viallet's, 1970; McRae, Suppan and microscopic solvent polarity parameter. ADS560EI dye is a very good media for efficient broadband dye laser in the blue-green region and fluorescing. The lasing properties of ADS560EI have led to a renewed interest in their spectral characteristics, which require complete and systematic study.

In continuation of our work (Raikar et al., 2010; 2006) on the determination of excited state, ground state dipole moments and rotation diffusion of some laser dyes in various solvents, we examined a new compound ADS560EI which constitute an important industrial application like security printing, lithographic printing plates and many other printing applications. In spite of many industrial applications, however, no systematic study related to excited state dipole moment with solvent effects and rotation diffusion studies has been reported until now. This prompted us to carry out the present work. The main aim of the present work is to estimate and

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compare the ground and excited state dipole moment of ADS560EI by various methods and to determine the rotation diffusion of the said molecule.

At present, there is no perfect theory that can correlate all the known solute and solvent interactions. It is very difficult to describe the solvatochromic shifts completely. However the determination of the ground and excited state dipole moments of dye molecule is important because the values of dipole moment provide information about the change in electronic distribution upon excitation. The nature of excited states reflects the charge distribution in the molecule and allows one to judge the site of attack by electrophilic and nucleophilic reagents in some photochemical reactions. To estimate the short lived dipole moments many methods are used such as electronic polarization of fluorescence, electronic dichroism, stark splitting of rotational levels, microwave conductivity and thermochromic shift. However, their use is limited because of intensive equipment and restriction to relatively small molecule. The solvatochromic method (Bilal et al., (2001); Thipperudrappa et al., (2008)) is based on a linear correlation between the wave numbers of the absorption and fluorescence maxima and a solvent polarity function which involves both dielectric constant (ϵ) and refractive index (n) of the surrounding medium.

EXPERIMENTAL

Apparatus

Electronic UV/VIS absorption measurements are made using Hitachi 150-20 spectrophotometer and fluorescent measurements are on Hitachi F2000 spectrofluorometer at room temperature.

Fluorescence life times were measured using an IBH (Scotland, UK) fluorescence spectrometer based on Time-Correlated-Single-Photon-Counting (TCPSC) technique. The sample was excited by laser with pulse width of ~100ps. The fluorescence was detected by PMT based deflection module (Pal et al., (1990)) (model TBX4, IBH, Scotland, UK) placed at right angles to the excitation path.

Chemicals

AR-Grade Laser Dye ADS560EI was obtained from American dye source, Inc, Canada and used without further purification. The molecular structure and IUPAC name is given in Figure 1. All the solvents viz., methanol, ethanol, propan-2-ol, 1-octanol, butanol, nananol, ethyl acetate, acetonitrile, DMSO, benzene, toluene and glycerol are of spectroscopic grade.

General Procedure

Determination of Dipole moment

The independent equations are used for the determination of ground and excited state dipole moments are as follows

Lippert's Equation

$$\overline{V}_a - \overline{V}_f = m_1 F(\epsilon, n) + const \quad (1)$$

Bakhshiev's equation

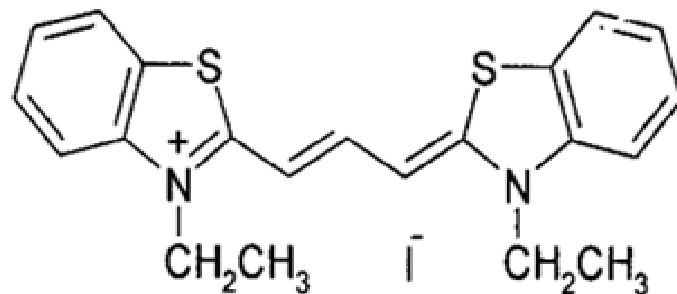


Figure 1. Molecular Structure of ADS560EI with IUPAC name. 3-Ethyl-2-[3-[3-ethyl-3H-benzothiazol-2-ylidene]-1-propenyl]-benzothiazolium iodide

$$\overline{V}_a - \overline{V}_f = m_2 F_1(\epsilon, n) + const \quad (2)$$

Kawski-Chamma-Viallet's equation

$$\frac{\overline{V}_a + \overline{V}_f}{2} = -m_3 F_2(\epsilon, n) + const \quad (3)$$

McRae's Equation

$$\overline{V}_a = -m_4 F_3(\epsilon) + const \quad (4)$$

Suppan's equation

$$\overline{V}_a = -m_5 F_4(\epsilon) + const \quad (5)$$

The expression $F(\epsilon, n)$ [Lippert's polarity parameter], $F_1(\epsilon, n)$ [Bakhshiev's polarity parameter], $F_2(\epsilon, n)$ [Kawski-Chamma-Viallet's polarity parameter], $F_3(\epsilon)$ [McRae's polarity parameter] and $F_4(\epsilon)$ [Suppan's polarity parameter] are given as:

$$F(\epsilon, n) = \left[\frac{\epsilon - 1}{2\epsilon + 1} - \frac{n^2 - 1}{2n^2 + 1} \right] \quad (6)$$

$$F_1(\epsilon, n) = \frac{2n^2 + 1}{n^2 + 2} \left[\frac{\epsilon - 1}{\epsilon + 2} - \frac{n^2 - 1}{n^2 + 2} \right] \quad (7)$$

$$F_2(\epsilon, n) = \left[\frac{2n^2 + 1}{2(n^2 + 2)} \left(\frac{\epsilon - 1}{\epsilon + 2} - \frac{n^2 - 1}{n^2 + 2} \right) + \frac{3(n^4 - 1)}{2(n^2 - 1)^2} \right] \quad (8)$$

$$F_3(\epsilon) = \left[\frac{2(\epsilon - 1)}{\epsilon + 2} \right] \quad (9)$$

$$F_4(\epsilon) = \left[\frac{2(\epsilon - 1)}{2\epsilon + 2} \right] \quad (10)$$

Where \overline{V}_a and \overline{V}_f are absorption and fluorescence maxima are

in wave numbers (cm^{-1}) respectively. \mathcal{E} and n are dielectric constant and refractive index of the solvents respectively. Plotting the graphs of Equations 1-5 it follows that $(\bar{\nu}_a - \bar{\nu}_f)$ versus $F_2(\mathcal{E}, n)$, $(\bar{\nu}_a - \bar{\nu}_f)$ versus $F_1(\mathcal{E}, n)$, $\frac{1}{2}(\bar{\nu}_a + \bar{\nu}_f)$ versus $F_2(\mathcal{E}, n)$, $\bar{\nu}_a$ versus $F_3(\mathcal{E})$ and $\bar{\nu}_a$ versus $F_4(\mathcal{E})$ should give linear graphs with slopes m_1, m_2, m_3, m_4, m_5 respectively and these are given as:

$$m_1 = \frac{2(\mu_e - \mu_g)^2}{hca^3} \quad (11)$$

$$m_2 = \frac{2(\mu_e - \mu_g)^2}{hca^3} \quad (12)$$

$$m_3 = \frac{2(\mu_e^2 - \mu_g^2)}{hca^3} \quad (13)$$

$$m_4 = \frac{\mu_g(\mu_e - \mu_g)}{hca^3} \quad (14)$$

$$\text{And } m_5 = \frac{\mu_g(\mu_e - \mu_g)}{hca^3} \quad (15)$$

Where μ_g and μ_e are the ground and excited state dipole moments of the solute molecule respectively, h and c are plank's constant and velocity of light in vacuum respectively and a is the Onsager cavity radius determined by the atomic increment method (Edward, 1956), assuming the fact that ground and excited state dipole moments are almost parallel to each other. The following expressions are obtained on the basis of Equations 12 and 13.

$$\mu_g = \frac{m_3 - m_2}{2} \left[\frac{hca^3}{2m_2} \right]^{1/2} \quad (16)$$

$$\mu_e = \frac{m_3 + m_2}{2} \left[\frac{hca^3}{2m_2} \right]^{1/2} \quad (17)$$

And

$$\mu_e = \left[\frac{m_2 + m_3}{m_3 - m_2} \right] \mu_g \quad (18)$$

The validity of these equations is based on certain assumptions:

1. μ_g and μ_e dipole moments are collinear.
2. The Onsager cavity radius is same in ground and excited state.

3. Polarizability of the molecule is ignored.
4. Hydrogen bonding effect is also ignored.

These theoretical calculations are also compared by correlation as the spectral shift with E_T^N parameter proposed by Reichardt et al. (1988) and improved by Ravi et al. (1995).

$$\bar{\nu}_a - \bar{\nu}_f = 11307 \cdot 6 \left[\left(\frac{\delta\mu}{\delta\mu_B} \right)^2 \left(\frac{a_B}{a} \right)^3 \right] E_T^N + \text{const} \quad (19)$$

Where $\delta\mu_B = 9\text{D}$ and $a_B = 6.2\text{\AA}$ for are dipole moment changes on excitation and Onsager cavity radius respectively of a bitanine dye molecule (Reichardt et al., 1988). $\delta\mu$ and a are corresponding quantities of ADS560EI laser dye of our present interest. The change in dipole moment can be evaluated from the slope of Stokes shift versus E_T^N plot and is given by the equation.

$$\Delta\mu = \mu_e - \mu_g = \sqrt{\frac{m \times 81}{(6.2/a^3) \times 11307 \cdot 6}} \quad (20)$$

Determination of Reorientation time

Steady state reorientation time of Laser Dye ADS560EI is measured using steady state depolarization method in methanol solvent of spectroscopic grade. The steady state anisotropy $\langle r \rangle$ is related to experimentally measured intensities by the following equation.

$$\langle r \rangle = \frac{I_{\parallel} - I_{\perp}}{I_{\parallel} + 2G I_{\perp}} \quad (21)$$

Where I_{\parallel} and I_{\perp} are polarized fluorescence intensities parallel and perpendicular with respect to the excitation radiation. G is an instrumental factor which corrects the polarization bias in the detection system and is given by:

$$G = \frac{I_{HV}}{I_{HH}} \quad (22)$$

Where I_{HV} is the fluorescence intensity in which the excitation polarizer is kept horizontal and the emission polarizer vertical and I_{HH} is the fluorescence intensity when the both the polarizer are kept horizontal.

The sample was excited at 556nm and emission was monitored from 574nm to 589nm. The measurement of $\langle r \rangle$ involves recording four spectra each for I_{\parallel} and I_{\perp} and two for the G -factor. Each anisotropy measurement was repeated 5-6 times and for every trial the G -factor was determined. The experiment was performed 298K. Reorientation time can be obtained from the measured steady state anisotropies by the following Perrin relation. The decay of fluorescence and the decay of anisotropy are single exponential [Lackowich JR (1983)].

$$\tau_r = \frac{\tau_f}{\left[\left(\frac{r_0}{\langle r \rangle} \right) - 1 \right]} \quad (23)$$

Where r_0 , τ_f and τ_r

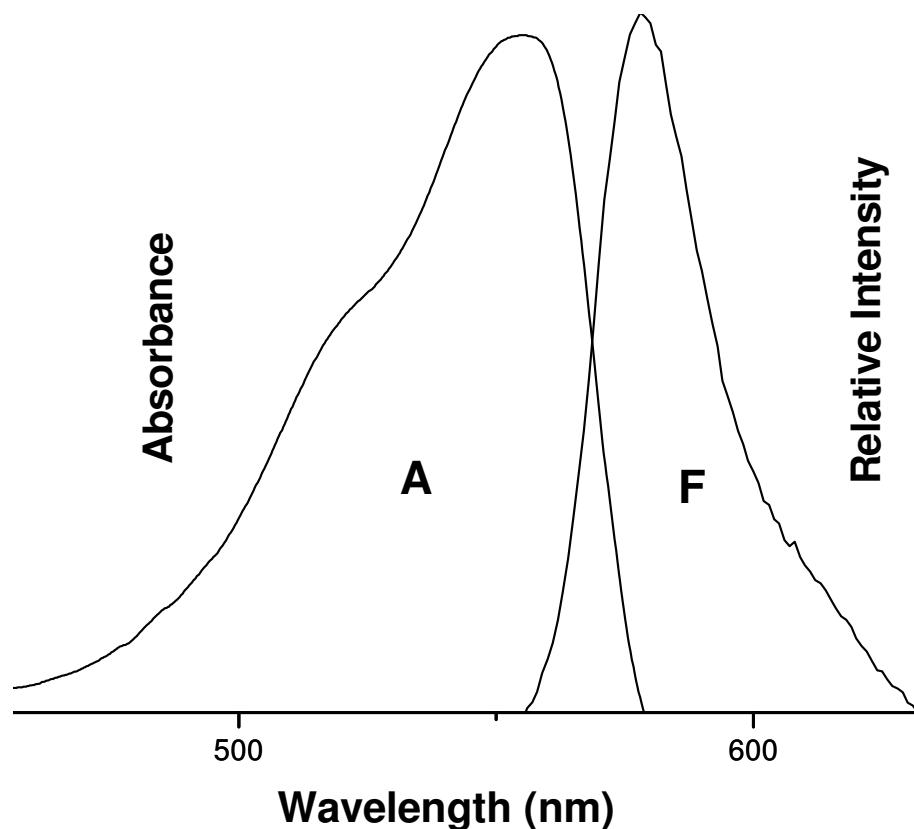


Figure 2. Absorption and fluorescence spectra of ADS560EI in Ethanol.

are limiting anisotropy, fluorescence life time and reorientation time respectively. The limiting anisotropy r_0 value was determined by measuring the steady state anisotropies of the probe in glycerol at low temperature (below -15°C). As glycerol has high viscosity in this condition, all rotational motions are frozen.

RESULTS AND DISCUSSION

Absorption and fluorescence emission spectra of a ADS560EI molecule is recorded in various solvents of different solvent parameters of dielectric constant (ϵ) and refractive index (n) of various solvents. Figure 2 shows the typical absorption and fluorescence spectra of a molecule in ethanol solvent. The values of Stokes shift are indicated in Tables 1 and 2, absorption and maximum emission is indication of charge transfer during excitation. Larger magnitude of the Stokes shift indicates that the excited state geometry could be different from that of ground state. The general observation is that there is an increase in the Stokes shift values when there is an increase in the solvent polarity, which shows that there is an increase in the dipole moment on excitation. The fluorescence emission peaks undergoes a red shift, confirming a $\pi \rightarrow \pi^*$ transition. Figure 3-7 show the graphs of $(\bar{\nu}_a - \bar{\nu}_f)$ versus $F(\epsilon, n)$, $(\bar{\nu}_a - \bar{\nu}_f)$ versus

$F_1(\epsilon, n)$, $\frac{1}{2}(\bar{\nu}_a + \bar{\nu}_f)$ versus $F_2(\epsilon, n)$, $(\bar{\nu}_a - \bar{\nu}_f)$ versus $F_3(\epsilon)$, $(\bar{\nu}_a - \bar{\nu}_f)$ versus $F_4(\epsilon)$ from which slopes m_1, m_2, m_3, m_4, m_5 respectively are obtained. The plot of $(\bar{\nu}_a - \bar{\nu}_f)$ versus E_T^N gives Figure 8 the slope value m . The slopes, intercepts and correlation coefficients for these fitted graphs are given in Table 3.

The ground state dipole moment (μ_g) of the ADS560EI laser dye is estimated by using Equation 16 and is presented in Table 4. However, no other experimental data on μ_g could be obtained from literature for comparison. The μ_e Values obtained from Equations 11-15, 17 and 20, and the ratio (μ_e / μ_g) obtained from Equation 18 are also presented in Table 4. The differences between the values of μ_e may be due to the various assumptions and simplifications made in the use of different Solvatochromic methods. The value μ_e obtained by Lippert method is large, since it does not consider the polarizability. The μ_e values obtained by Suppan method is large compared to all

Table 1. Wave numbers (cm^{-1}) for the absorption and fluorescence emission maxima of ADS560EI molecule in different solvents and Stokes shift.

Solvent	Absorption max (nm)	Fluorescence max (nm)	$\bar{\nu}_a$	$\bar{\nu}_f$	$(\bar{\nu}_a - \bar{\nu}_f)$	$\frac{1}{2}(\bar{\nu}_a + \bar{\nu}_f)$	E_T^N
Methanol	556.4	578.6	17972.68	17283.09	689.59	17627.89	0.762
Ethanol	558.8	582.0	17895.49	17182.13	713.36	17538.81	0.654
Propanol	560.4	574.2	17844.39	17415.53	428.86	17629.97	0.546
Butanol	561.6	577.8	17806.27	17307.03	499.24	17556.65	0.543
Octanol	564.4	578.2	17717.93	17295.05	422.88	17506.49	0.602
Nonanol	564.8	578.0	17705.38	17301.04	404.34	17503.21	--
Ethyl acetate	562.6	580.4	17774.62	17229.49	545.12	17502.06	0.228
Acetonitrile	554.8	572.4	18024.51	17470.30	554.21	17747.41	0.460
DMSO	564.4	585.5	17717.93	17079.42	638.51	17398.67	--
Benzene	578.0	589.6	17301.04	16960.65	340.39	17130.84	0.111
Toluene	576.4	577.8	17349.06	17307.03	42.04	17328.04	0.099
Glycerol	562.2	584.2	17787.26	17117.43	669.84	17452.34	0.812

Table 2. Dielectric constant and refractive index of various solvents and Lippert's, Bakshiev's, Kowski-Chamma-Viallet's, McRae's and Suppan's polarity parameters.

Solvent	ϵ	n	$F(\epsilon, n)$	$F_1(\epsilon, n)$	$F_2(\epsilon, n)$	$F_3(\epsilon)$	$F_4(\epsilon)$
Methanol	33.00	1.328	0.31	0.86	0.65	1.83	0.95
Ethanol	25.30	1.361	0.29	0.82	0.65	1.78	0.94
Propanol	20.18	1.377	0.28	0.78	0.65	1.73	0.93
Butanol	17.84	1.399	0.26	0.75	0.65	1.70	0.92
Octanol	10.30	1.429	0.23	0.63	0.60	1.51	0.86
Nonanol	8.83	1.427	0.21	0.57	0.57	1.45	0.84
Ethyl acetate	6.02	1.372	0.20	0.49	0.50	1.25	0.77
Acetonitrile	38.80	1.344	0.31	0.87	0.67	1.85	0.96
DMSO	46.50	1.479	0.26	0.84	0.74	1.88	0.97
Benzene	2.28	1.499	3.24×10^{-3}	7.04×10^{-3}	0.34	0.60	0.46
Toluene	2.38	1.497	0.01	0.03	0.35	0.63	0.48
Glycerol	46.53	1.472	0.26	0.84	0.74	1.88	0.97

other methods. The μ_e values obtained by McRae and E_T^N method almost agree with one another. The change in the dipole moment between the excited state and ground state is due to intramolecular charge transfer (ICT). Further, a large change in dipole moment on excitation suggests that excited state is twisted intramolecular charge transfer (TICT). Presence of a large TICT and increase in polarity on excitation render the molecule more polar (as compared to ground state) giving rise to a large change in the dipole. In literature one may find that large number of investigators have used Solvatochromic shift method to estimate and excited state dipole moments. In literature there is dearth of data on μ_g and μ_e for the ADS560EI molecule used in the present study. Therefore, we have not attempted to make any comparison between the different methods.

Figure 9 gives a plot of τ_r versus η in various solvents. From Table 5, it can be seen that there is linear relationship between τ_r (ps) and viscosity (mPa). The reorientation times of ADS560EI is large in Nonanol that is experiences more friction than in the methanol solvent. This can be explained by considering the interaction of alcohol molecules with probe molecule, which results from hydrogen bonding. The cavities formed in this network of solvent molecules have become regions of reduced friction and hence the molecule could rotate faster in methanol than the nonanol.

Conclusions

We have estimated and compared the excited state dipole moment of ADS560EI laser dye molecule as a function of solute-solvent interactions by various methods using

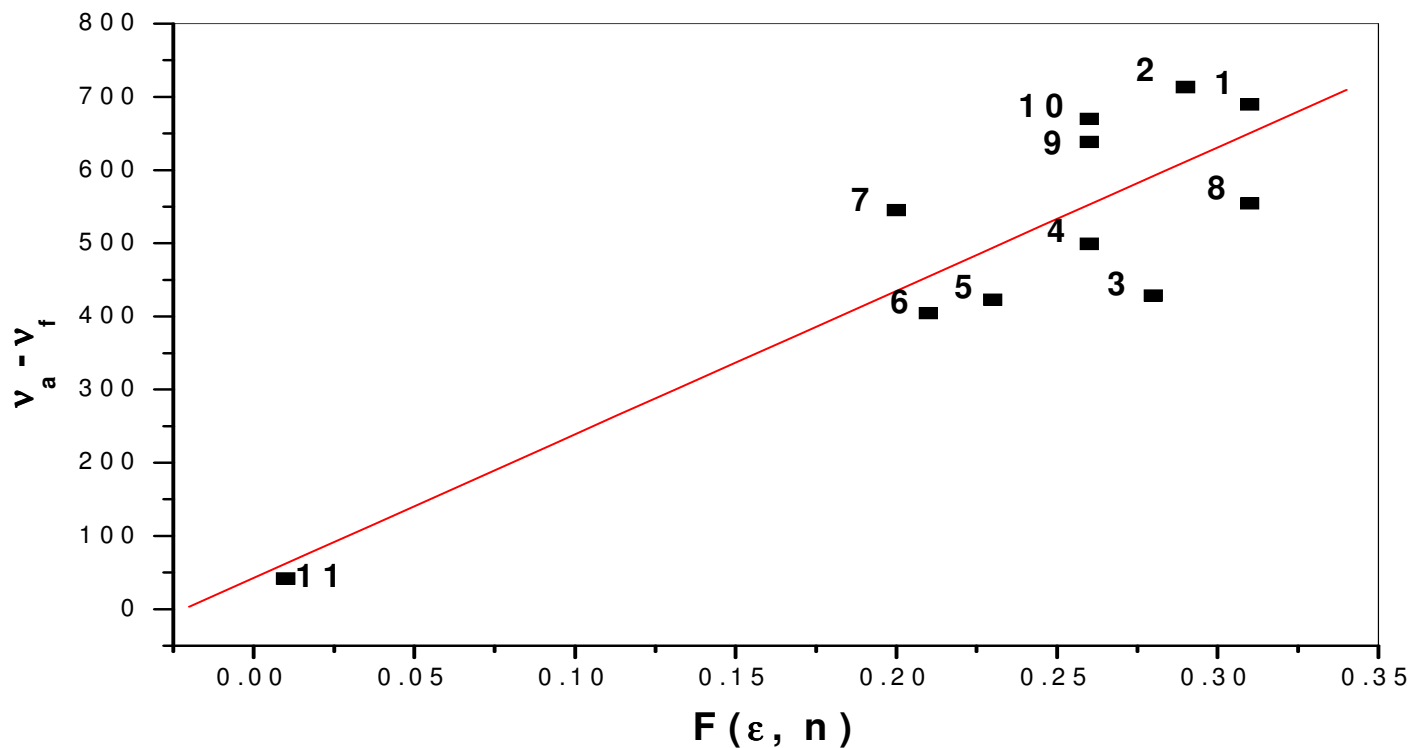


Figure 3. Plot of Stokes shift versus $F(\epsilon, n)$ that is Lippert's Polarity parameter of ADS560EI in different solvents. (1) methanol, (2) ethanol, (3) propanol, (4) butanol, (5) octanol, (6) nananol, (7) ethylacetate, (8) acetonitrile, (9) DMSO, (10) toluene, (11) glycerol. (-----Linear fit).

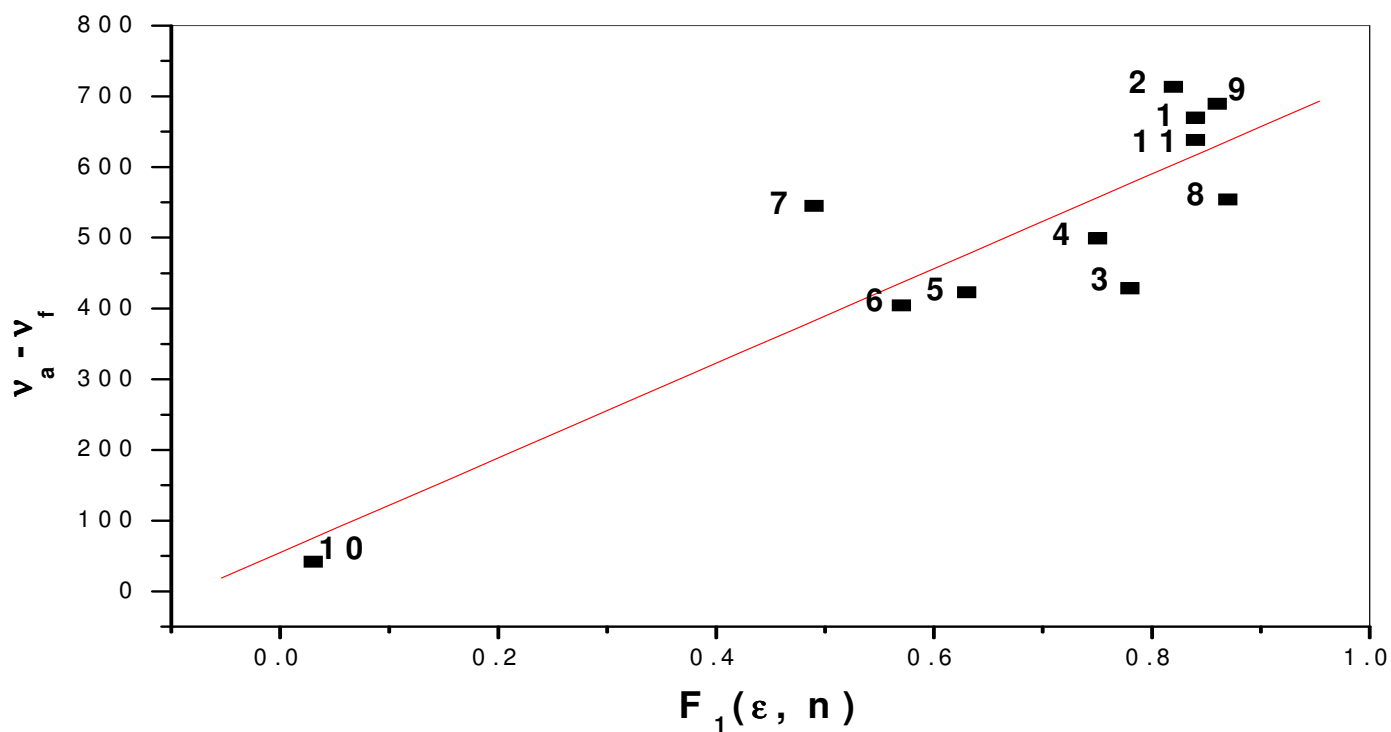


Figure 4. Plot of Stokes shift versus $F_1(\epsilon, n)$ that is Bakshiev's Polarity parameter of ADS560EI in different solvents. (1) methanol, (2) ethanol, (3) propanol, (4) butanol, (5) octanol, (6) nananol, (7) ethylacetate, (8) acetonitrile, (9) DMSO, (10) toluene, (11) glycerol (-----Linear fit).

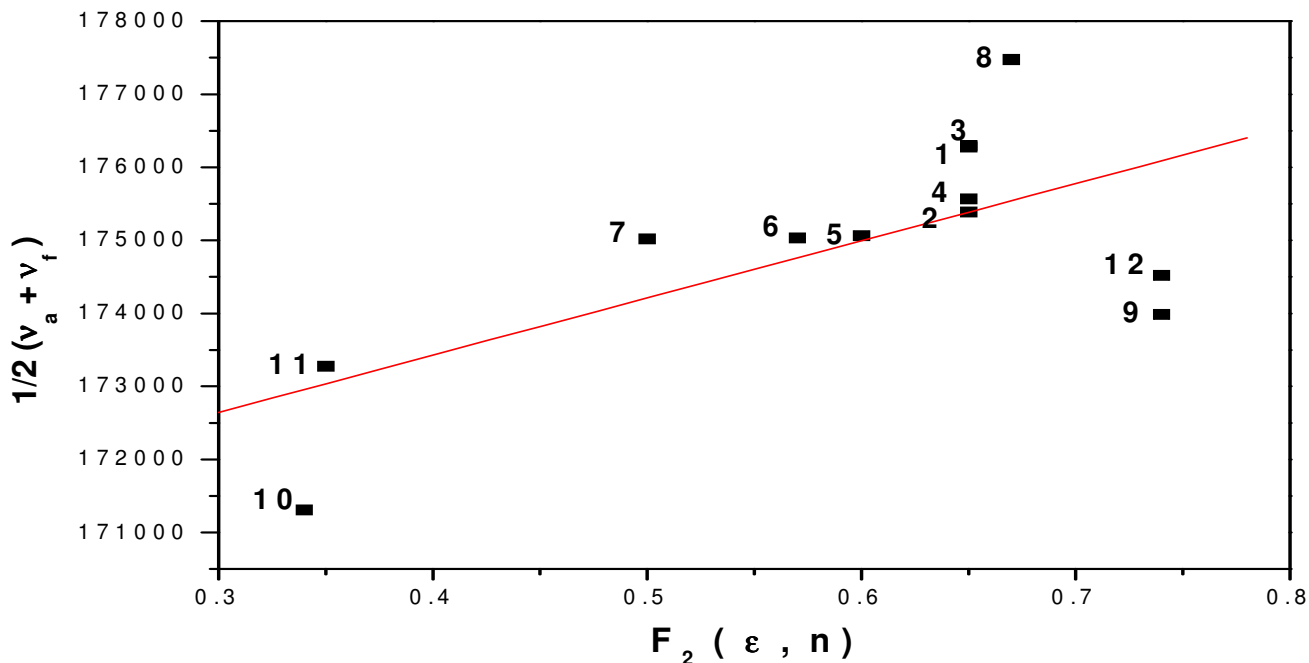


Figure 5. Plot of $\frac{1}{2}(\overline{v_a + v_f})$ versus $F_2(\epsilon, n)$ that is Kawski-Chamma-Viallet's Polarity parameter of ADS560EI in different solvents. (1) methanol, (2) ethanol, (3) propanol, (4) butanol, (5) octanol, (6) nananol, (7) ethylacetate, (8) acetonitrile, (9) DMSO, (10) benzene, (11) toluene, (12) glycerol, (-----) Linear fit).

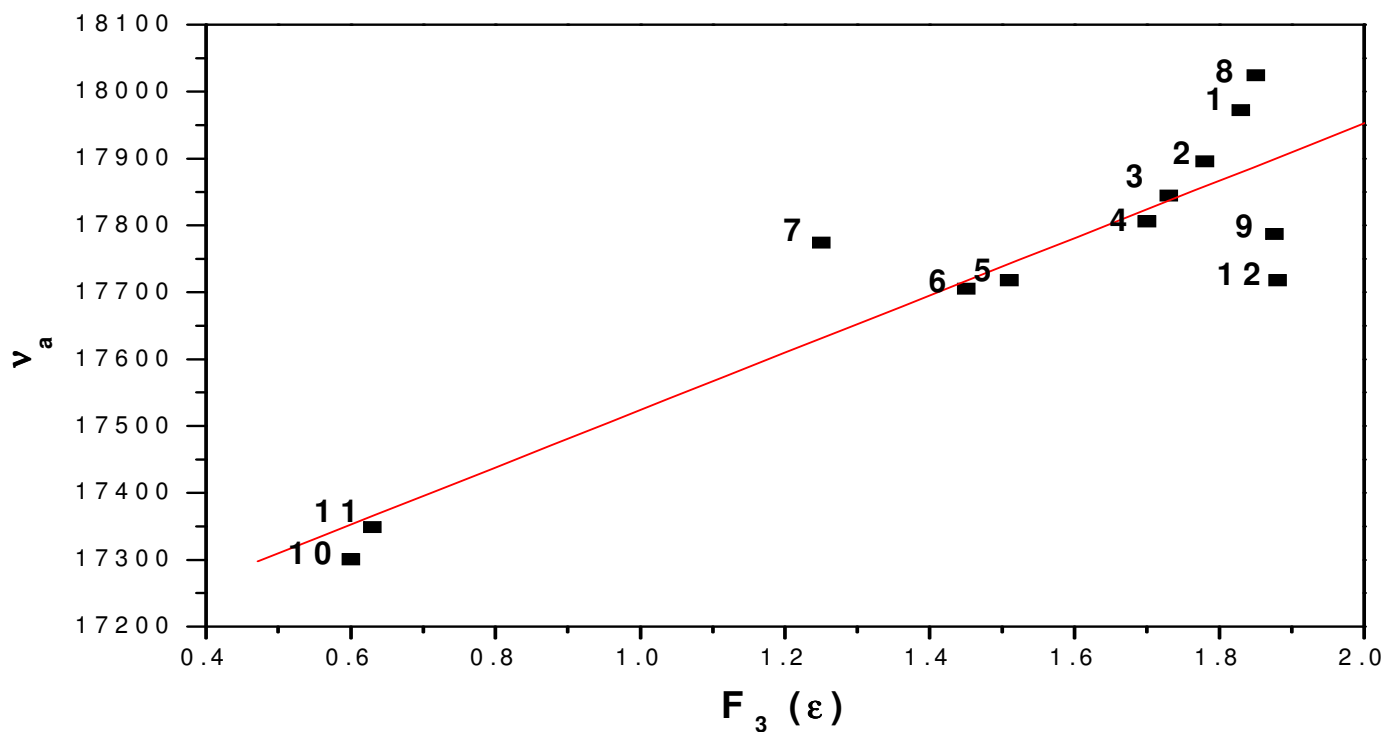


Figure 6. Plot of $\overline{v_a}$ versus $F_3(\epsilon)$ that is McRae's Polarity parameter of ADS560EI in different solvents. (1) methanol, (2) ethanol, (3) propanol, (4) butanol, (5) octanol, (6) nananol, (7) ethylacetate, (8) acetonitrile, (9) DMSO, (10) benzene, (11) toluene, (12) glycerol, (-----) Linear fit).

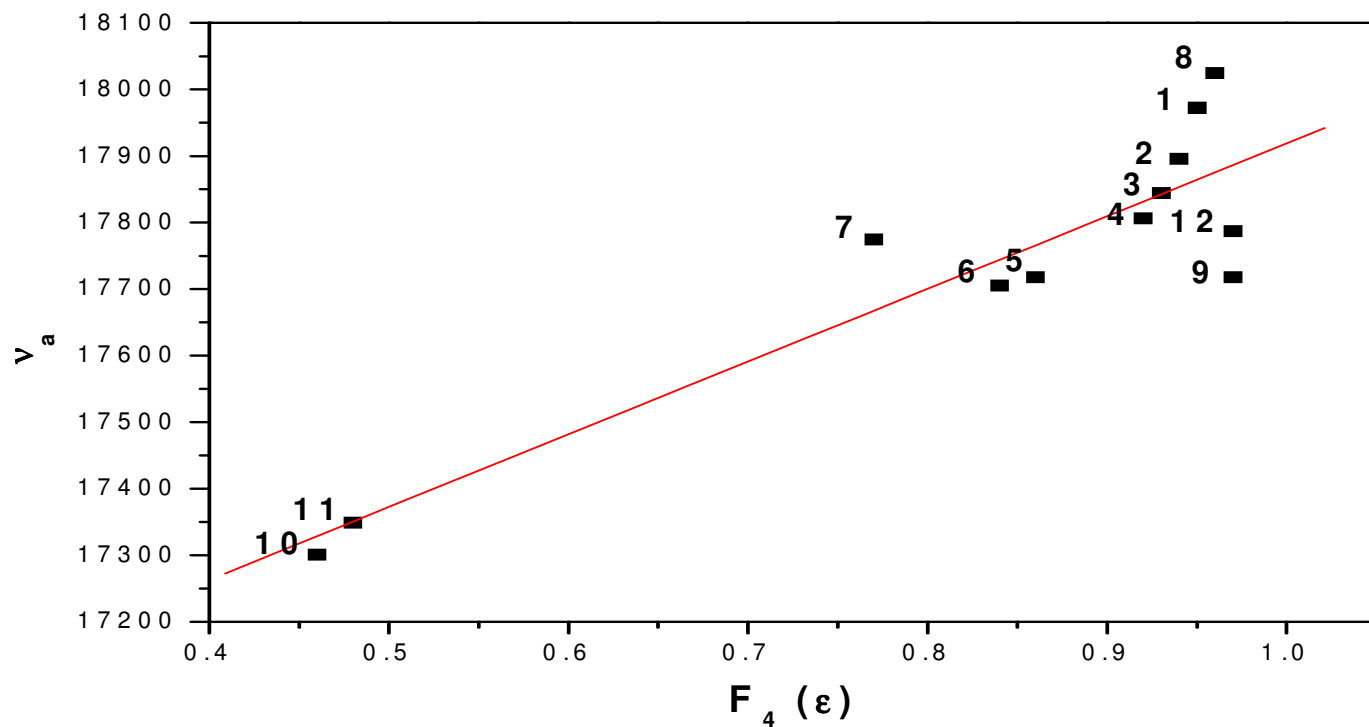


Figure 7. Plot of $\bar{\nu}_a$ versus $F_4(\epsilon)$ that is Suppan's Polarity parameter ADS560EI in different solvents. (1) methanol, (2) ethanol, (3) propanol, (4) butanol, (5) octanol, (6) nananol, (7)ethylacetate, (8) acetonitrile, (9)DMSO, (10) benzene (11) toluene, (12) glycerol, (-----Linear fit).

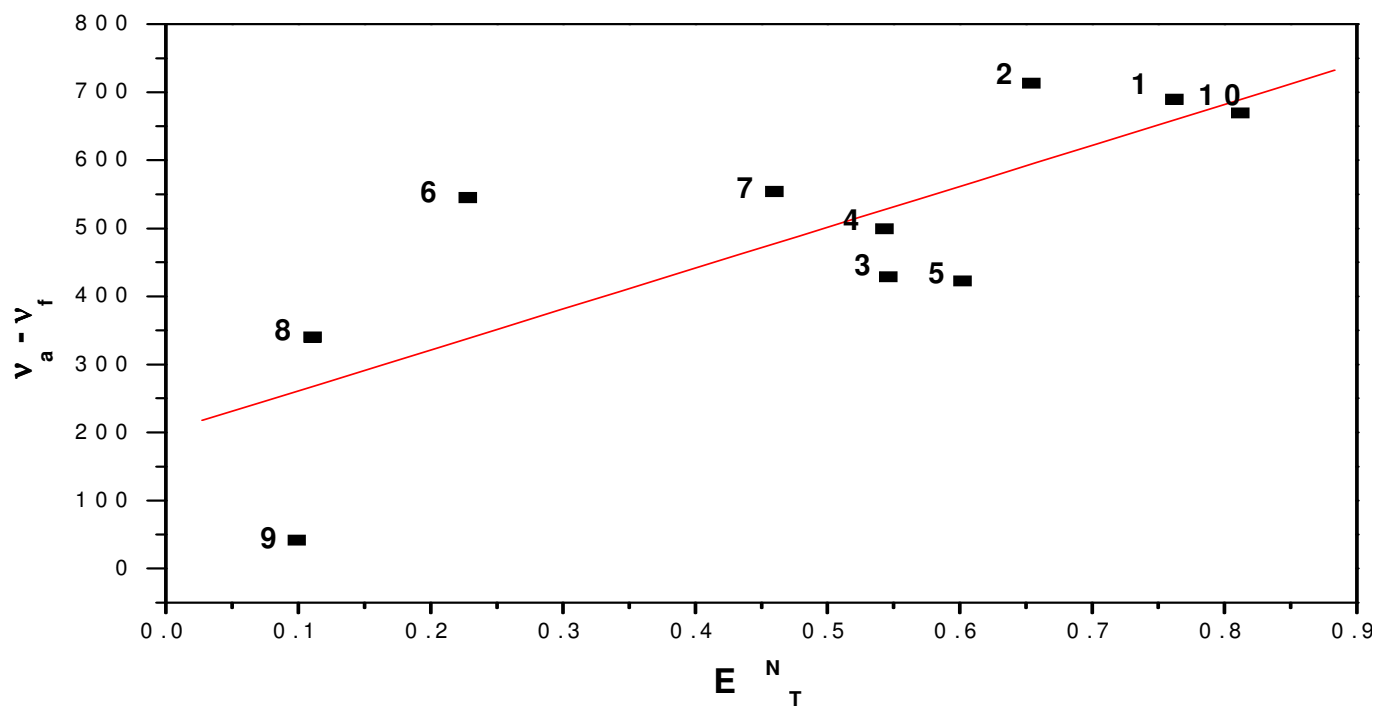


Figure 8. Plot of Stokes shift versus E_T^N that is Microscopic solvent Polarity parameter ADS560EI in different solvents. (1) methanol, (2) ethanol, (3)propanol, (4) butanol, (5)octanol, (6)ethylacetate, (7) acetonitrile, (8)benzene (9)toluene, (10)glycerol, (-----Linear fit)

Table 3. Statistical data's of the correlations of the spectral shifts of ADS560EI laser dye.

Method	Slope	Intercept	Correlation coefficient	No. of data
Lippert's	19614.09	426.45	0.864	12
Bakhshiev's	6693.44	546.63	0.880	12
Kawski Chamma Viallet's	7828.89	170296.70	0.656	12
McRae's	428.67	17095.29	0.901	12
Suppan's	1093.27	16825.77	0.910	12
E_T^N	601.45	200.84	0.768	09

Table 4. Onsager cavity radius, Ground state and excited state dipole moment (in Debye, D) results of ADS560EI Laser Dye by various methods.

Onsager cavity radius 'a' (Å)	μ_g^a	μ_e^b	μ_e^c	μ_e^d	μ_e^f	μ_e^h	μ_e / μ_g^i
4.37	0.61	7.81	12.92	11.47	28.32	12.50	12.78

^a Ground state dipole moments calculated from Equation (16), ^bExcited state dipole moments calculated from Equation (17) Solvatochromic method, ^cExcited state dipole moments calculated from Lippert's equation Equation (11), ^dExcited state dipole moments calculated from Mc Rae's equation Eq. (14), ^f Excited state dipole moments calculated from Suppan's equation Equation (15), ^h Excited state dipole moments calculated from microscopic solvent polarity function (20), ⁱRatio of excited to ground state dipole moments calculated from Equation (18).

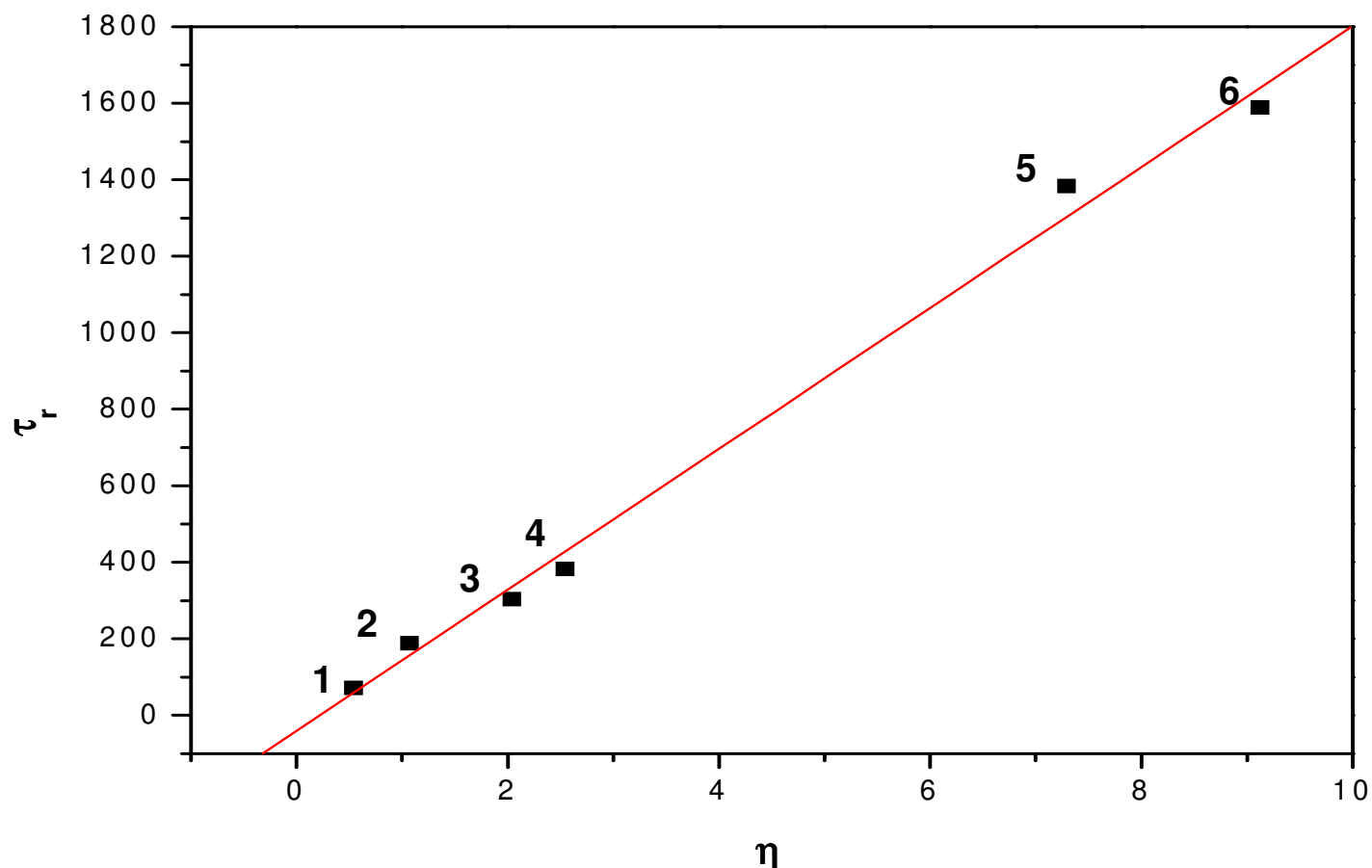
**Figure 9.** Plot of reorientation time (τ_r) versus viscosity of ADS560EI in different solvents. (1)methanol, (2) ethanol, (3)propanol, (4) butanol, (5)octanol, (6) nananol, (-----)Linear fit).

Table 5. Fluorescence life time of ADS560EI molecule in a various solvents.

Solvents	η (mPa)	$\langle r \rangle$	τ_f /ns	τ_r (ps)
Methanol	0.54	0.141	0.099	72
Ethanol	1.07	0.158	0.21	189
Propanol	2.04	0.177	0.27	304
Butanol	2.54	0.190	0.29	383
Octanol	7.29	0.233	0.60	1384
Nananol	9.12	0.236	0.66	1589

Stokes shift. It can be seen that the dipole moment in the excited state by different methods ranges between 7.81 to 28.32 D. The large difference between the excited state and ground state dipole moment is due to the intra molecular charge transfer and twisted intra molecular charge transfer. Also, we have estimated the reorientation time of a molecule in various solvents, the larger the viscosity of the solvent molecule, the slower the rotation of the probe molecule or the larger the reorientation time. To the best of our knowledge this is the first report on the dipole moment and reorientation time calculation for ADS560EI molecule and would be of great help in many fields as mentioned in the introduction.

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