study of the optical properties for BDN-I dye solutions which are used to Q-switch the Nd:YAG laser

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Abstract:
The Nd:YAG laser system has been mounted and the BDN-I dye was prepared and dissolved in different pure organic solvents such as (carbon-tetra-chloride, chloroform, Acetone, Dioxane, and pyridine) and in a mixture of carbon-tetra-chloride with another polar organic solvent at 1:1 and 2:1 mixing ratios. The dye solution was situated in a dye cell, and modulated inside the laser resonator. The giant laser pulse power, and the dye solution transmittance (T) have been measured. The (T) values are utilized in calculation of the optical properties such as (Reflectance (R), Absorptance (A) and refractance (r)), some indices as optical density (d), refractive index (n), relative refractive index (n_r). The effect of the variation of the BDN-I molar concentration dye solution and mixing ratios has been studied on the optical properties firstly, the effect of the optical properties on each other secondly, and finally, the effects of the BDN-I dye solutions optical properties on the giant laser pulse characteristics. It can be concluded that the relative refractive index (n_r) has an important role in controlling the passive Q-switching process by specifying the optical properties of the BDN-I solution. The main conclusion that the mixing method of carbon-tetra-chloride with other organic solvent at different mixing ratios is the best way to improve the giant laser pulse characteristics, where it decreases the optical properties which causes the losses of stimulated photons energy, and increases of that which causes consolidations the saturation absorption process.

1-Introduction:
Nd:YAG laser is a four-level solid-state laser which consists of Nd^{3+} in a crystal of Y_3Al_5O_{12} (the crystal being known as YAG) [1], where 1.0% Nd^{3+} ions substitutes for Y^{3+} ions [2]. The laser transition has (1.064 μm) wavelength (near-infrared) [1,2]. The rise time of the near-infrared fluorescence intensity in YAG:Nd^{3+} at room temperature under the excitation by (10nsec) duration.
pulses at (514.5 nm) has been measured by T.Kushida et al. [3]. The experimental and numerical study of the geometrical transfer efficiency of various cavities for Nd:YAG laser system are presented by I.Y.Milev et al. [4].

The Q-switching technique has been implemented on Nd:YAG laser by the saturable absorbers to achieve giant laser pulses which are more useful than which are obtained in free-running operation. There are many materials have been used as a passive Q-switches for Nd:YAG laser. These materials may be solids as (Cr,Nd) saturable absorber in (YAG) gain medium which are demonstrated a compact, efficient, highly polarized, and highly stable Q-switched laser [5-7]. Cr:YAG crystal has been performed as a saturable absorber for Nd:YAG laser [8-13].Cr^{4+}:GSGG saturable absorber [14], V:YAG crystal [15-16], or gases such as narrow-band of (CS₂) vapor saturable absorber [17]. The semiconductors could also be used as a Q-switches for Nd:YAG laser as InGaAs [18-19]. Many dyes which are dissolved in different solvents, used as a liquid Q-switches for Nd:YAG laser.

The nickel complex BDN-I is used as a saturable absorber for Q-switching neodymium lasers. It is particularly attractive for their good photochemical stability, the possibility of tailoring their ground-state recovery (GSR) time by the appropriate choice of solvent [20], where the optical properties of the BDN-I dye substantially differ depending on the host material [20], or the solvents which is dissolved in them [21-23].

BDN-I dye has the chemical name of Bis(4-dimethylaminodithiobenzil)nickel, and the molecular formula C₃₂H₃₀N₂NiS₄ (629.55) molecular weight, melting point of (270-280)°C [23].

The BDN-I dye was homemade and used to Q-switch the Nd:YAG laser at 1997 [24]. The study of using PMMA foils doped with mixture of BDN-I in different solvents as a solid Q-switches for Nd:YAG laser has been implemented experimentally and theoretically in 2001 [25], and the measurement of the threshold energy damage of them are achieved in 2003 [26]. A method of preparing complexes based on BDN-I as a Q-switches for the solid-state lasers, as, these complexes have resonance absorption bands near the emitted wavelength laser to produce saturable absorber dyes which are cover a wide range of resonance wavelengths, has been introduced in 2002 [27].

In this research well introduce a study of the optical properties of the BDN-I solution which consists of that dye dissolved in different pure organic solvents and in a mixture of the Carbon-tetra chloride with other solvent at 1:1 and 2:1 mixing ratios. The BDN-I solution is used as a passive Q-switch for the Nd:YAG laser at 1.064μm wavelength.

2-The optical properties of the laser passive Q-switch and the mechanism of the saturation absorption:

The saturable absorbers(S.A) are the materials (solids or liquids or gases) which can reveals a bleaching case after they are reach to the saturation absorption for incident photons on them. These materials are used as a passive Q-switches for the different kinds of lasers [28-30]. The passive Q-switching technique is an important way to generate a giant laser pulse which can be used in many applications [31-33]. This technique can be implemented by setting (S.A) inside the resonator, continuously pumping for an active medium, (S.A) molecules begin to absorb the incident photons energy and excited to higher excited states until the ground states become fully empty and the higher excited states completely filled with excited molecules, that means stopping the absorption, the (S.A) become fully transmitted to the stimulated photons incident on it, allowing to the accumulated population inversion to decay rapidly to the ground-state suddenly and generate a nanosecond duration, Megawatt power of the giant laser pulse [34-37].

The optical properties of the (S.A) act an important role in a saturation absorption mechanism where the (S.A) absorptance (A) is the molecules ability to absorb the stimulated photons energy and excite to higher energy levels, this is consolidate the Q-switching, while the losses of the stimulated photons by; withdrawing photons to the air medium in reflection process, them refraction inside the (S.A) (bending the array because of difference in optical density of (S.A) and that for air and photons transmission without any deviation because of vertically incident on the surface.
separates between (S.A), and the air [38-40]. The reflectance (R), refractance (r), and transmittance (T) of the (S.A) are obstruct the saturation absorption process and then lowest laser pulse power may be generated. Therefore, laser workers used materials with lowest R, r and T for chosen laser array and change many parameters in laser passive Q-switching system to moderate these properties with incident photons. These properties and the saturation absorption of the (S.A) are drawn in fig. (1 a, b, and c).

Fig.1:
(a)-The optical processes which are happened on the stimulated photons inside the (S.A).
(b)-The gradually saturation absorption of the stimulated photons inside the (S.A).
(c)- The occurring of the bleaching process in the (S.A). where
$I_i$: incident intensity, $I_A$: absorbed intensity
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\[ I_r : \text{refracted intensity}, \quad I_R : \text{reflected intensity} \]
\[ n' : \text{the air refractive index}, \quad n : \text{the (S.A) refractive index}. \]

3- Theory:

The refractive index of the BDN-I dye solution \( n \) which is related of \( T \), can be calculated as follow [41]:

\[ T = \frac{2n}{n^2 + 1} \quad \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots (1) \]

Eq(1) is the second order equation, so we can solved it by constitution way. It has two solutions which is illustrated as follow:

\[ n = \frac{2 + \sqrt{4 - 4T^2}}{2T} \quad \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots (2a) \]
\[ n = \frac{2 - \sqrt{4 - 4T^2}}{2T} \quad \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots (2b) \]

The reflectance \( R \) of the dye solution may be calculated as follow [41]:

\[ R = \left( \frac{n - 1}{n + 1} \right)^2 \quad \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots (3) \]

The other optical property is the refractance \( r \) of the (S.A) can be calculated as below [42]:

\[ r = \frac{I_i - I_r}{I_i} \quad \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots (4) \]

where \( I_r \) is the refracted beam intensity which is defined as below:

\[ I_r = n I_i \quad \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots (5) \]

and \( I_i \) is the incident beam intensity on the dye solution. Absorptance is the one of important optical properties which is equal to [42]:

\[ A = 1 - \log \left( \frac{1}{T} \right) \quad \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots (6) \]

The optical density \( d \) of the BDN-I dye solution can be calculated as follow [43]:

\[ d = \frac{\varepsilon c f}{\ell} \quad \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots (7) \]

where \( \varepsilon \) is the molar extinction coefficient of the (S.A) which is calculated by utilizing from Beer-Lambert law as below [42]:

\[ \varepsilon = \frac{1}{c f} \log \left( \frac{P_i}{P} \right) \quad \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots (8) \]

where \( c \) is the molar concentration of the BDN-I dye in solution, \( f \) is the cell thickness, \( P_i \) is the laser pulse power in free-running operation, and \( P \) is the Q-switched laser pulse power.

The relative refractive index \( n_r \) is the ratio of the refractive index of the dye solution \( n \) to that of air \( n' \) at the same wavelength of beam which is incident on the dye solution. \( n_r \) can be calculated as follow [44]:

\[ n_r = \frac{n \ ( \text{solution})}{n' \ ( \text{air})} \quad \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots (9) \]

4- Experimental part:

The system shown in fig.2 is used to measure the giant Nd:YAG laser pulse characteristics and the optical properties of the BDN-I dye solution which is used as a passive Q-switch for Nd:YAG laser. It consists of Nd:YAG rod (1) of (5cm) long and (5mm) diameter as an active medium with (54%) reflected mirror (2) fixed optically to its one end while the other end faces retrareflected prism (3) as a total reflected element.

The elliptical resonator of (12.6 cm) long consists of two halves. The Nd:YAG rod is mounted on one focus points and the flash lamp filled with Kr (4) is fixed in the second focus points. Using a pulse forming network with an external triggering (5) the flash lamp has been operated. Careful alignment of the free running cavity has been carried out using the He-Ne laser. An ED-200 genetic Joumlmeter which was supplied by (EG&G) company, has been used to measure the laser...
pulse energy. It has been connected to a fast oscilloscope (7) model (TDS 500) which was supplied by Tektronix company. The P-i-n detector (9) which was connected to oscilloscope is used to detect the transmitted photons from the dye cell (8). The cell is filled by BDN-I dye dissolved in one of each (CCl₄, CHCl₃, Acetone, dioxane, and Pyridine) pure solvent or in a mixture of (CCl₄) with one of each other solvents at 1:1 and 2:1 mixing ratios. It was putted firstly, inside the resonator (8) to measure the Q-switch pulse intensity (I) and secondly outside it (8’) to measure the incident intensity (Iᵢ) on it.

The beam splitter (expander) (10) is used to separate the photons array outside the resonator to two arrays in order to reduce the energy arrived to detector.

The incident intensity of the laser had been measured by removing the dye cell from the system, and we find that Iᵢ of (59.36 X 10⁻³ photons/cm².sec) intensity, but the transmitted intensity (Iₜ) is measured by putting the dye cell in (8’) position and making the calibration on the energy measurement appearing on the oscilloscope monitor. This procedure was repeated in each time of dissolving the BDN-I dye either in pure solvent or in a mixture of (CCl₄) with other solvent at 1:1 and 2:1 mixing ratios.

The Q-switched laser pulse characteristics (energy, duration, and power) were measured by system which is shown in fig.2 and putting the dye cell in (8) position.

![Diagram](image_url)

**Fig.2 :**
The system of the BDN-I dye solution optical properties measurement, where:
1. Nd:YAG rod.
2. 54% reflected mirror.
3. Retrareflected prism.
4. Flash lamp.
5. Flash lamp power supply and triggering system.
6. ED-200 genetic Joulmeter.
7. TDS 500 oscilloscope.
8. The dye cell inside the resonator.
8’. The dye cell outside the resonator.
9. P-i-n detector.

**5- The calculation and results :**
The optical properties of the BDN-I dye solution at 1.064 μm Nd:YAG laser wavelength such as (R, r, and A) had been calculated by eqs.(3,4, and 6), respectively. But the transmittance (T) of the BDN-I dye has been measured by ED-200 genetic Joulmeter at different values of molar concentration. The optical properties of the BDN-I dye which are dissolved in pure, and in a...
mixture of CCl$_4$ with other solvent at 1:1 and 2:1 mixing ratios, are depicted in figs.(3,4 and 5), respectively.

To interpret the optical properties of the BDN-I dye solution, we are calculated the optical density (d) by eq.(7), and the refractive index (n) by using the solution in eq.(2-b) and neglecting the other because it shows unsuitable value for the relative index of this dye solution. The (d) and (n) of the BDN-I dye dissolved in pure, or in a mixture of 1:1 and 2:1 mixing ratios, are depicted in fig.(6-a,b,c) and fig.(7-a,b,c), respectively.

The Q-switched laser pulse power (P) using BDN-I dye solution which are measured by the system shown in fig.(2), are listed in table (1-a,b,c).

The relative refractive index which is calculated according to eq.(9), is listed in table (2-a,b,c) for the BDN-I dye dissolved in pure, and in a mixture of 1:1 and 2:1 mixing ratios, respectively.

*Table(1)*:
The Q-switched Nd:YAG laser pulse power (P)* using BDN-I dye which is dissolved in: (a)- pure solvent. (b)- mixture of CCl$_4$ with other at 1:1 mixing ratio. (c)- mixture of CCl$_4$ with other at 2:1 mixing ratio. *PX10$^3$* watt for all tables.

<table>
<thead>
<tr>
<th>Cx10$^3$ mol/l</th>
<th>solvents</th>
<th>CCl$_4$</th>
<th>CHCl$_3$</th>
<th>C$_5$H$_5$N</th>
<th>C$_3$H$_6$O</th>
<th>C$_4$H$_8$O$_2$</th>
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<td>9.92</td>
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<td>-</td>
<td>4.48</td>
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</table>
-b-

<table>
<thead>
<tr>
<th>Cx10⁻³ mol/l</th>
<th>( \text{CCl}_4 + \text{C}_2\text{H}_8\text{O}_2 )</th>
<th>( \text{CCl}_4 + \text{C}_5\text{H}_5\text{N} )</th>
<th>( \text{CCl}_4 + \text{CHCl}_3 )</th>
<th>( \text{CCl}_4 + \text{C}_3\text{H}_6\text{O} )</th>
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<td>( 0.7 )</td>
<td>3.97</td>
<td>1.70</td>
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<tr>
<td>( 0.8 )</td>
<td>3.84</td>
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<td>3.74</td>
<td>1.68</td>
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</table>

\( \text{Mixtures} \)

-c-

<table>
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<th>Cx10⁻³ mol/l</th>
<th>( \text{CCl}_4 + \text{C}_5\text{H}_5\text{N} )</th>
<th>( \text{CCl}_4 + \text{CHCl}_3 )</th>
<th>( \text{CCl}_4 + \text{C}_3\text{H}_6\text{O} )</th>
<th>( \text{CCl}_4 + \text{C}_4\text{H}_8\text{O}_2 )</th>
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<td>( 0.9 )</td>
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</table>

\( \text{Mixtures} \)

**Table (2):**
The relative refractive index \( (n_r) \) of the BDN-I dye solution at 1.064 \( \mu \text{m} \) Nd:YAG laser wavelength, when this dye is dissolved in:

(a)- pure solvents.
(b)- mixture of \( \text{CCl}_4 \) with other solvent at 1:1 mixing ratio.
(c)- mixture of \( \text{CCl}_4 \) with other solvent at 2:1 mixing ratio.
### Table 1: Solvents Mixtures

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<tr>
<th>Cx10³ mol/l</th>
<th>CCl₄</th>
<th>CHCl₃</th>
<th>C₂H₅N</th>
<th>C₃H₆O</th>
<th>C₄H₈O₂</th>
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### Table 2: Mixtures

<table>
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<tr>
<th>Cx10³ mol/l</th>
<th>CCl₄ + CHCl₃</th>
<th>CCl₄ + C₂H₅N</th>
<th>CCl₄ + C₃H₆O</th>
<th>CCl₄ + C₄H₈O₂</th>
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Discussion:

Figures (3-5 a,b,c,d) show the optical properties of the BDN-I dye solutions as (transmittance (T), reflectance (R), refractance (r), and absorbance (A)) as a functions of the dye molar concentration (C). The study of (C) variation effects on T, R, r, and A for BDN-I dye which is dissolved in pure organic solvent such as (CCl₄, CHCl₃, C₃H₆O, C₄H₈O₂, and C₅H₅N), and in mixtures of (CCl₄) with another organic solvent at 1:1 and 2:1 mixing ratios, are shown in figures (3-5 a,b,c,d), respectively. We observed all optical properties of pyridine solution are drawn alone in single figure as shown in fig. (3, e) because of the large difference in (C) values which were appear the Q-switching phenomena. All these figures are explained; the decreasing of (T) and (A), increasing of (R) and (r) at the same increasing values of molar concentration (C). The increasing of (C) makes the dye molecules are crowded in ground-states, so the large amount of incident stimulated photons on the dye cell is absorbed by the molecules, then they have been excited to the higher excited states, all that are interpreted the (A) decreasing with (C) increasing, where:

$$A = (I_i - I_A) / I_i$$

The highest dye molecules density has the role in rebounding part of the incident stimulated photons to the air (medium which is coming from it) due to the increasing of its optical density (d) with increasing of (C) as shown in fig (6,a,b,c,d), that is interpreted the increasing of (R) with (C) from one side, and from another side the high number of molecules in the ground-state are contributed in stimulated photons deviation from them path inside the dye solution that is (in other meaning) increasing (r) because of the refractive index increasing as illustrated in fig (7,a,b,c,d).

The largest (T) and (A) of (0.644) and (0.378), respectively are obtained when BDN-I dye dissolved in (CCl₄) solvent at (9.92 x 10⁻³ mol/l). The values of (T) and (A) gradually to less values from above when BDN-I dissolved in solvents as dioxane, then chloroform, then Acetone, lately pyridine at the same value of (C). We observed the decreasing of (T) and (A) when dye dissolving in a mixture of (CCl₄) with another solvent at 1:1 mixing ratio, and less values to mixture of 2:1 from the values to a pure solvents. They refers to dilution ratios increasing by (CCl₄) causes decreasing of (T) and (A) for BDN-I dye solution at Nd:YAG laser wavelength. That is attributed to the solvent which has highest polarity, contribute in making the BDN-I dye solution more transmitted and absorbed than from less polarity solvent, as shown in appendix (1). Except of pyridine which show less transmittance from Acetone, in spite of it's has more polarity than Acetone because of the effective atom in pyridine molecule (N)
which has electro negativity of ( 3.5) less than for Oxigine (O) atom of (3) electro negativity which is the effective atom in Acetone molecule.

The giant laser pulse generation with highest power has been specified by firstly , less absorptance which is nearly equal to zero , that is near all incident photons are fully absorbed by dye molecules , ( A ) may be reached to a negative values that is exactly indicator for found of other photons in the laboratory , which is contributes in dye absorption . Secondly, too low ( T ) values which means that the large amount of incident photons are transmitted without any deviation at the end of the saturation absorption process . All the power values for all the used solvents ( pure or mixture at 1:1 and 2:1 mixing ratio ) are listed in table ( 1 ) .

At the low molar concentration ( C ) value , the BDN-I dye solution has less reflectance ( R ) and refractance ( r ) , and caused less power of the Q-switched laser pulse ; because of the highest refractive index ( n ) of the dye solution at low ( C ) . The ( r ) values may be nearly equal to zero or has low values , that refers to approach the refractive index of the dye solution ( n ) to that for air ( n' ) . In other wards , the relative refractive index ( n_r ) nearly equal to ( 1 ) at lowest BDN-I dye molar concentration ( C ) .Table (2,a,b,c,) show all the relative refractive index of the dye solutions at different ( C ) .

**Conclusion:**

When we want to design a passive laser Q-switching system by liquid dye , we must choose a solution which has more ( R ) and ( r ) , and less ( T ) and ( A ) in order to ensure the best giant laser pulse . We can depend on the dye molar concentration increasing in realization these optical properties ( R , r , T , and A ) , the relative refractive index ( n_r ) of the BDN-I dye solution also has the effective rule in generation of the best Q-switched laser pulse characteristics by its controlling on refractance ( r ) and reflectance ( R ) in two different cases , first , when ( n_r ) was in lowest value , ( r ) , ( R ) and pulse power have the highest value . Second , when ( n_r ) approach to ( 1 ) , (r) , ( R ) and ( P ) decay gradually . The polarity of the solvent specifies the transmittance and absorptance of the BDN-I dye solution . It can be concluded that the pyridine solvent contributes in generation of high power giant laser pulse when it used to dissolve the BDN-I dye , either pure or a mixture of 1:1 and 2:1 mixing ratios , because of its hasing highest optical properties with lowest molar concentration of the dye . We can consider that the mixing method of the solvents is the best way to generate high power Q-switched laser pulse .

**Appendix (1):**
The polarity of the solvents which are used to Q-switch the Nd:YAG laser [45].

<table>
<thead>
<tr>
<th>solvents</th>
<th>Temperature(C°)</th>
<th>Polarity X10^{-24} (cm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CCl₄</td>
<td>18</td>
<td>1.2, 0.9, 1.2</td>
</tr>
<tr>
<td>C₄H₈O₂</td>
<td>40</td>
<td>1.1</td>
</tr>
<tr>
<td>CHCl₃</td>
<td>20</td>
<td>1.6</td>
</tr>
<tr>
<td>C₅H₆N</td>
<td>30</td>
<td>1.7, 3.4</td>
</tr>
<tr>
<td>C₃H₈O</td>
<td>40</td>
<td>1.4, 1.3, 3.4, 7.33</td>
</tr>
</tbody>
</table>

**Appendix (2):**
The electro negativity of the effective atoms in the solvents molecules which are used to dissolve the BDN-I dye as a liquid passive Q-switch for Nd:YAG laser [46].
<table>
<thead>
<tr>
<th>Element</th>
<th>Electro negativity</th>
</tr>
</thead>
<tbody>
<tr>
<td>O</td>
<td>3.0</td>
</tr>
<tr>
<td>N</td>
<td>3.0</td>
</tr>
<tr>
<td>Cl</td>
<td>3.0</td>
</tr>
<tr>
<td>C</td>
<td>2.0</td>
</tr>
<tr>
<td>H</td>
<td>2.1</td>
</tr>
</tbody>
</table>
Fig (3)
The effects of the molar concentration variations for BDN-I dye dissolved in pure organic solvents on its optical properties toward Nd:YAG laser array where:
(a) - The relation of Transmittance with molar concentration for four different solvents.
(b) - The relation of Reflectance with molar concentration for four different solvents.
(c) - The relation of Refrectance with molar concentration for four different solvents.
(d) - The relation of Absorptance with molar concentration for four different solvents.
(e) - The relations of all optical properties with molar concentration for Pyridine solvent only.
Reflectance

(b) Molar Concentration X 10^{-3} mol/l

Reflectance

(c) Molar Concentration X 10^{-3} mol/l
The effects of the molar concentration variations for BDN-I dye dissolved in a mixture of carbon-tetra chloride with other organic solvent in 1:1 ratio on its optical properties toward Nd:YAG laser array where:

(a) The relation of Transmittance with molar concentration for four different mixtures.
(b) The relation of Reflectance with molar concentration for four different mixtures.
(c) The relation of Refractance with molar concentration for four different mixtures.
(d) The relation of Absorptance with molar concentration for four different mixtures.
Fig(5)
The effects of the molar concentration variations for BDN-I dye dissolved in a mixtures of carbon-tetra chloride with other organic solvent in 2:1 ratio on its optical properties toward Nd:YAG laser array where:

(a) - The relation of Transmittance with molar concentration for four different mixtures.
(b) - The relation of Reflectance with molar concentration for four different mixtures.
(c) - The relation of Refrectance with molar concentration for four different mixtures.
(d) - The relation of Absorptance with molar concentration for four different mixtures.
The effects of the molar concentration variations for BDN-I dye on its optical density toward Nd:YAG laser array when it dissolved in:

(a)- four different organic solvents.
(b)- a mixtures of Carbon –tetra chloride with other organic solvents in 1:1 ratio.
(c)- a mixtures of Carbon –tetra chloride with other organic solvents in 2:1 ratio.
(d)- a Pyridine solvent only.
The effects of the molar concentration variations for BDN-I dye on its refractive index toward Nd:YAG laser array when it dissolved in

(a) four different organic solvents
(b) a mixtures of Carbon –tetra chloride with other organic solvents in 1:1 ratio.
(c) a mixtures of Carbon –tetra chloride with other organic solvents in 2:1 ratio.
(d) a Pyridine solvent only.

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