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GLASS LASER SYSTEMS

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REPORT NO. 82
OCTOBER 1978

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We present physical properties data for a number of liquids currently being used for cooling and/or index matching of components in high peak power, low repetition rate Nd: glass laser systems. Liquids examined include saturated water solutions of $ZnCl_2$ and $ZnBr_2$, TCP, benzene, DMSO, glycerol, ethylene glycol, FC-104, Dow-Corning 710, and Cargille 5040 and 5610. Thermal properties (conductivity, diffusivity), mechanical properties (viscosity, density), chemical properties (pH, materials compatibility), and optical properties (refractive index, Abbé value, nonlinear index, dn/dT , optical transparency) are tabulated and compared to projected liquid properties requirements for high repetition rate systems.

Key words: Abbé value; coolants, dn/dT ; index matching liquids; Neodymium glass lasers; nonlinear refractive index; thermal conductivity; viscosity.

1. Introduction

A variety of liquids serve as important components of current moderate repetition rate glass laser systems. Specifically, these liquids find application as coolants and as index matching mediums in amplifiers and electro-optic isolation systems. In addition, future systems may have other uses for liquids such as in liquid electrodes for electro-optic devices.

It is the intent of this paper to provide data on the physical properties which are important for the selection of the most appropriate liquid for a given application. In what follows we identify the materials currently in use or proposed for use in large glass laser systems, and the physical properties of interest. We then give a description of the various measurement techniques utilized, followed by a tabulation of data. Finally, a discussion of results including advantages/disadvantages of certain liquids in particular applications is presented.

2. Liquids

A dozen liquids were chosen for inclusion in this study. These liquids are identified by manufacturer, grade, lot number, and use in table 1.

Table 1. Liquids of interest

Liquid	Manufacturer	Grade/Lot#	Description
Distilled H_2O			Coolant
Ethylene glycol	Mallinckrodt	AR/EGK	Coolant
Tricresyl phosphate	J. T. Baker	515303	Index match - from mixed cresols
Glycerol	Mallinckrodt	AR/WRDN	Index match
Dimethyl sulfoxide	Eastman	Reagent/B44	Index match
Benzene	Fisher	AR/714259	Index match
$ZnBr_2$ (aq)	Cargille	optical/551	Index match
$ZnCl_2$ (aq)			Index match - prepared @ LLE with Fisher solid $ZnCl_2$ AR/746575
DC-710	Dow-Corning	BFC345	Index match - silicone oil
C5040	Cargille	"ND 1.495 @ 10,600Å"	Index match - immersion liquid, hydro-generated terphenyl and mineral oil
C5610	Cargille	"ND 1.495 @ 10,600Å"	Index match - immersion liquid, silicone oil
FC-104	3M	526	Fluorochemical flashtube coolant

$ZnBr_2$ and $ZnCl_2$ are both highly acidic and corrosive; they do however exhibit stability under intense UV flashlamp irradiation. The Cargille liquids 5040 and 5610 can be obtained from the manufacturer

mixed to a desired refractive index of between 1.46 and 1.56. These Cargille liquids possess an extremely low water content, and are resistant to high voltage breakdown, making them compatible with KD*P in Pockels cell devices. Liquids such as benzene, tricresyl phosphate, and dimethyl sulfoxide pose health hazards [1]¹.

3. Measurements and apparatus

The physical properties discussed in this paper fall into four categories: optical, thermal, thermo-optic, and mechanical. Specific properties studied include the following: refractive index at $\lambda = 1.064\mu\text{m}$, $0.6328\mu\text{m}$, $0.6563\mu\text{m}(n_c)$, $0.5876\mu\text{m}(n_d)$, $0.4861\mu\text{m}(n_F)$; Abbé value v_d ; spectral transmittance from $0.2\mu\text{m}$ to $1.4\mu\text{m}$; absorption coefficients at a variety of wavelengths; a calculated value for n_2 the nonlinear refractive index; the temperature coefficient of refractive index dn/dT ; thermal conductivity, density and viscosity. Table 2 gives a listing of measurement apparatus and experimental precision obtained for the above.

Table 2. Measurement apparatus and precision

Measurement	Apparatus/Technique	Precision
Refractive index	Hilger-Chance Refractometer [2]	± 0.0005
Abbé value, v_d	"	$\pm 7\%$ low Abbe values $\pm 50\%$ high Abbe values
Nonlinear refractive index, n_2	"	$\pm 8\%$ large values n_2 $\pm 50\%$ small values n_2
Absorption coefficient α	Cary 14 spectrophotometer	$\pm 0.015 \text{ cm}^{-1}$
Spectral Transmittance	"	
Temp. coeff. of ref. index, dn/dT	Minimum angle of deviation	$\pm 5\%$
Thermal conductivity, k @ 25°C	Thermal comparator [3]	$\pm 3\%$
Density, ρ @ 38°C	Gay-Lussac pycnometers	$\pm 0.003 \text{ gm/ml}$
Viscosity, kinematic, ν @ 21°C	Cannon-Fenske viscometers	ν , $\pm 5\%$
Viscosity, absolute, V @ 38°C	ASTM sizes 25, 100, 200, 450	V , $\pm 3\%$

Refractive index is determined using a Hilger-Chance refractometer [2], constructed of BK-7 and calibrated with Hoya BSC-7 glass. The device has a range of 1.25-2.0. Yag, HeNe, and dye laser sources are used to provide the wavelengths necessary to generate the index data.

The Abbé value, v_d is calculated from the standard formula,

$$v_d = \frac{n_d - 1}{n_F - n_C} \quad (1)$$

Figure 1 gives a plot of n_d versus v_d .

The electronic part of the nonlinear refractive index, n_2^e , which causes self-focusing of optical beams [4], is calculated using v_d , n_d and the following relationship:

$$n_2^e (10^{-13} \text{ esu}) = \frac{68(n_d - 1)(n_d^2 + 2)^2}{v_d \left\{ 1.517 + \left[(n_d^2 + 2)(n_d + 1)v_d \right] / 6n_d \right\}^2} \quad [5] \quad (2)$$

Figure 2 gives a plot of n_d versus n_2 .

A Cary spectrophotometer provides the spectral transmittance of each liquid from $0.2\mu\text{m}$ to $1.4\mu\text{m}$. Absorption coefficient data is similarly obtained from this instrument. For spectral scans the samples are contained in a quartz cell with an optical pathlength of 3.7 cm. Measurements are made at a scanning rate of 50Å/sec. These scans are presented in figures 3-8. Absorption coefficients have been calculated for five wavelengths ($1.34\mu\text{m}$, $1.064\mu\text{m}$, $1.054\mu\text{m}$, $0.6328\mu\text{m}$, $0.3550\mu\text{m}$) using the relationship

¹Figures in brackets indicate the literature references at the end of this paper.

$$\alpha_{\text{cm}^{-1}} = \frac{(\text{optical density}) + 2 \log_{10}(1-R_1) + 2 \log_{10}(1-R_2)}{L(\log_{10}e)} \quad (3)$$

where L = length of cell = 1.0 cm

$$R_1 = \text{reflectivity - quartz/air interface} = \left(\frac{n_{\text{quartz}} - 1}{n_{\text{quartz}} + 1} \right)^2$$

$$R_2 = \text{reflectivity - quartz/liquid interface} = \left(\frac{n_{\text{quartz}} - n_{\text{liquid}}}{n_{\text{quartz}} + n_{\text{liquid}}} \right)^2$$

n = refractive index at appropriate wavelength.

The temperature coefficient of refractive index, dn/dT , is measured via a minimum angle of deviation technique at $\lambda = 1.06\mu\text{m}$ near room temperature. The liquid is placed in a hollow glass prism (vertex angle $30^\circ 50'$); the deflection of a laser beam passing through the liquid at minimum angle of deviation is determined as the liquid temperature is changed from 29°C to 23°C . Temperature is monitored by a thermocouple placed within a capillary tube inside the prism. The value of dn/dT is calculated from the following equation

$$\frac{dn}{dT} = \frac{\cos \frac{1}{2}(\alpha + \theta)}{2 \sin \frac{1}{2} \alpha} \frac{d\theta}{dT} \quad (4)$$

where α = calibrated angle of prism = $30^\circ 50'$

$$\theta = 2 \sin^{-1}(n_{1.064} \sin \alpha/2) - \alpha$$

$$\frac{d\theta}{dT} = \frac{(\text{beam displacement in mm})}{(\text{path length in mm}) (\Delta T)}$$

ΔT = temperature change in $^\circ\text{C}$

Beam displacements are typically 10-20 mm for path lengths of 15-18 meters.

Thermal conductivity, important for heat flow calculations, is measured at room temperature for each liquid with a thermal comparator [3]. This instrument records the rate of cooling of the tip of a probe heated to 9°C above ambient upon contact with the teflon membrane of a liquid sample cell. A thermocouple pair responds to the temperature change by generating a microvolt signal which can be converted to thermal conductivity by calibration with reference standards. Figure 9 gives a thermal conductivity plot established with such standards. Thermal conductivity values for the samples are determined from this calibration curve with the microvolt signals obtained.

Density at 38°C is determined per ASTM D369-67 using Gay-Lussac type pycnometers. The filled pycnometers are heated to $38^\circ\text{C} \pm 0.1^\circ\text{C}$ in a water bath whose temperature is controlled using a Thermomix 1460 [6]. Mass determinations are made with a Sartorius 2474 analytical balance [7]. The volume of the pycnometers is obtained using a standard value for the density of H_2O at 38°C [8].

Viscosity at 21°C and 38°C is determined using Cannon-Fenske type viscometers. Each viscometer comes with a factory calibration at 38°C and is experimentally calibrated at 21°C . Temperature is maintained at 38°C using the Thermomix and at 21°C by using a large water bath at room temperature. The kinematic viscosity, ν in centistokes, is found as $\nu = Ct$ where C is the calibration constant of the viscometer and t is the efflux time of the travel of the liquid meniscus under the pull of gravity between two etched marks on a capillary tube. Absolute viscosity, V in centipoise, is expressed as $V = \nu\rho$ where ρ is the density of the liquid at the appropriate temperature. The absolute viscosity is determined at 38°C using density values measured at 38°C and the kinematic viscosity found with the viscometer calibration at this temperature. Kinematic viscosity is determined at 21°C only, because no density data was determined at this temperature.

4. Results

The data generated in this investigation are presented in three different forms. Table 3 collects numerical data on the physical properties of interest. Figures 1, 2, and 9 present graphs of n_d versus ν_d , n_d versus n_2 and thermal conductivity versus thermal comparator signal respectively. Figures 3-8 give spectral transmittance scans of the liquids.

Measured values of refractive index range from approximately 1.56 (TCP) to 1.27 (FC-104). The Hilger-Chance technique results in measured values of refractive index in close agreement with published values (generally ± 0.001). Table 4 lists a number of liquids and compares our data with published refractive index values.

Table 3. Physical properties of liquids for high repetition rate glass laser systems

Property	Water	Ethylene Glycol	Tricresyl Phosphate	Glycerol	Dimethyl Sulfoxide	Benzene	ZnBr ₂	ZnCl ₂	Dow-Corning 710	Cargille 5610	Cargille 5040	3M FC-104
Optical												
n_D Refractive index, $\lambda=1.06\mu\text{m}$	1.327	1.424	1.539	1.466	1.468	1.487	1.538	1.523	1.517	1.496	1.498	1.271
n_C $\lambda=.6563\mu\text{m}$	1.333	1.430	1.552	1.472	1.477	1.497	1.551	1.532	1.528	1.506	1.506	1.272
n_A $\lambda=.6328\mu\text{m}$	1.333	1.430	1.551	1.472	1.476	1.498	1.552	1.534	1.529	1.508	1.506	1.273
n_d $\lambda=.5876\mu\text{m}$	1.334	1.432	1.556	1.474	1.479	1.502	1.555	1.536	1.533	1.510	1.511	1.273
n_F $\lambda=.4861\mu\text{m}$	1.338	1.437	1.569	1.480	1.487	1.513	1.567	1.545	1.544	1.520	1.519	1.275
v_F Abbé value	67	62	33	59	48	31	35	41	33	36	39	91
n_2 Calculated nonlinear index (10^{-13} -esu)	.6	.9	3.6	1.1	1.6	3.1	3.2	2.4	3.2	2.7	2.3	.25
$\alpha_{1.34\mu\text{m}}$ Absorption coeff. cm^{-1}	5.029	.669	.128	.738	.105	.162	1.208	1.013	.093	.093	.104	.005*
$\alpha_{1.064\mu\text{m}}$	.153	.205	.029	.242	.046	.028	.091	.080	.023	.023	.051	.006*
$\alpha_{1.054\mu\text{m}}$	.162	.214	.026	.254	.035	.028	.102	.087	.027	.023	.057	.006*
$\alpha_{.6328\mu\text{m}}$	.014*	.019	.015*	.040	.015*	.017	.020	.021	.021	.021	.021	.013*
$\alpha_{.3550\mu\text{m}}$	.005*	.007*	.222	.040	.017	.016	.107	.145	.327	.231	2.503	.006*
Approx. UV edge μm (0.0-1.0 for 3.7cm cell)	<.200	.210	.345	.250	.280	.280	.290	.280	.330	.330	.375	.210
Thermo-Optic												
dn/dt Temp. coeff. ref. index (10^{-4})/°C	-1.2	-2.7	-3.3	-2.2	-3.6	-5.7	-2.6	-2.5	-3.1	-3.1	-3.6	-3.8
Thermal												
K, thermal conductivity cal/cm-sec-°C(10^{-3})	1.45	.52	.26	.38	.34	.35	.59	.45	.24	.22	.24	.23
Mechanical												
ρ Density @ 38 °C gm/cm ³	.99299 [8]	1.101	1.152	1.250	1.082	.859	2.447	2.097	1.089	1.049	.897	1.727
Viscosity @ 38 °C, centipoise	.69	10.23	30.0	320	1.62	.52	15.22	81.9	248	89.5	10.74	1.02
Kinematic viscosity @ 21 °C, centistokes	.98	15.8	62	915	1.97	.74	10.5	101	566	156	21.0	.77

*->99% transmission

Table 4. Refractive index measurements compared to literature values

Liquid	Measured values	Values from other sources
H ₂ O	$n_C = 1.333$ $n_D = 1.334$ $n_F = 1.338$	$n_C = 1.331$ [9] $n_D = 1.333$ [9] $n_F = 1.337$ [9] $n_D = 1.333$ [10]
Ethylene glycol	$n_D = 1.432$	$n_D = 1.432$ [10] $n_D = 1.429$ [8]
Tricresyl phosphate	$n_D = 1.556$	$n_D = 1.556$ [9]
Benzene	$n_D = 1.502$	$n_D = 1.501$ [9] $n_D = 1.501$ [10] $n_D = 1.498$ [8]
Glycerol	$n_D = 1.474$	$n_D = 1.473$ [9]
FC-104	$n_{1.06} = 1.271$ $n_C = 1.272$ $n_{.6328} = 1.273$ $n_D = 1.273$	1.271, λ unspecified [11]
DC-710	$n_D = 1.533$	1.533, λ unspecified [12]

The Abbé value which was calculated using refractive index data requires some explanation. It is seen that large Abbé values have errors of approximately $\pm 50\%$ while small Abbé values have calculated errors of $\pm 7\%$. Because a high Abbé value indicates low dispersion while a low Abbé value indicates large dispersion; it follows that for low dispersion as seen in FC-104 and water, the limit of $n_F - n_C$ as $n_F + n_C = 0$. Hence, v_D is large and any small change in n_F , n_C , or n_D can have a large effect on v_D .

The absorption coefficients and the spectral transmittance scans indicate wavelength regions in which transmission through thick samples of certain liquids would not be advisable due to strong absorption losses. Liquids exhibiting the strongest IR absorption are water, aqueous $ZnCl_2$ and $ZnBr_2$, ethylene glycol and glycerol. All the test liquids are highly transparent in the visible. Differences in absorption coefficients at $.3550\mu m$ can be attributed mainly to the effect of the UV edge for the different liquids as indicated in table 3. The importance of the UV edge in relation to flashlamp degradation will be discussed later. It is noteworthy that H₂O and ethylene glycol which absorb in the IR near $1.06\mu m$ also have the lowest UV edges (with FC-104) of the liquids tested.

The temperature coefficient of refractive index for water, $1.34 \times 10^{-4}/^\circ C$ is clearly substantially lower than dn/dT for any other liquid tested. The measured dn/dT values for liquids are approximately 10^2 times larger in magnitude than dn/dT values for glasses at $1.06\mu m$ [13]. All the liquids exhibit negative dn/dT values in that the refractive index decreases as the liquid temperature increases. From a product information sheet on Cargille immersion liquids [14], dn/dT values for 15-35°C average $-3.3 \times 10^{-4}/^\circ C$ which are close to measured values for C5040 and C5610 of $-3.6 \times 10^{-4}/^\circ C$ and $-3.1 \times 10^{-4}/^\circ C$ respectively.

Thermal conductivity values fall between $1.45 \text{ cal/cm-sec-}^\circ C \times 10^{-3}$ for water to approximately $.25 \text{ cal/cm-sec-}^\circ C \times 10^{-3}$ for TCP, DC-710, C5040, C5610 and FC-104. The plot in figure 9 of absolute signal versus thermal conductivity is a least squares fit of the data for calibration standards CCl_4 , toluene, ethanol, methanol, and H₂O [3]. This plot has a coefficient of determination $r^2 = .96$. Little conductivity data is available for liquids other than the calibration standards. Product literature gives conductivity data for FC-104 and DC-710. FC-104 has a reported conductivity of $.15 \text{ cal/cm-sec-}^\circ C \times 10^{-3}$ [11] versus a measured value of $.23 \text{ cal/cm-sec-}^\circ C \times 10^{-3}$. For DC-710 the published value is $.35 \text{ cal/cm-sec-}^\circ C \times 10^{-3}$ [12]; the measured value is $.24 \text{ cal/cm-sec-}^\circ C \times 10^{-3}$. For these two liquids

the percentage difference between published and measured values is large. It should be noted however, that ZnBr_2 and ethylene glycol were measured at two different times using the same technique with resultant conductivity values within the stated precision of $\pm 3\%$.

Measured viscosity and density exhibit a wide range of values. Viscosity is low for H_2O , benzene, FC-104 and DMSO; it is two orders of magnitude higher for DC-710 and glycerol. Density values range from approximately .9 gm/ml for benzene and C5040 to 2.4 for ZnBr_2 . It is interesting to note that benzene and FC-104 have kinematic viscosities which are practically identical but density differences result in the absolute viscosity of FC-104 being twice that of benzene at 38°C .

There is generally good agreement between measured and published values of viscosity and density. Table 5 compares those values for water, benzene, glycerol, ethylene glycol, FC-104 and DC-710.

Table 5. Measured values of density and viscosity compared to other sources

Liquid	Measured values	Data from other sources
Water	$V_{38} = .69\text{cp}$	$\rho_{38} = .99299 \text{ gm/ml}$ [8]
	$v_{21} = .98\text{cs}$	$V_{38} = .678\text{cp}$ [8]
		$V_{38} = .681\text{cp}$ [9]
		$v_{21}^* = .980\text{cs}$ [8]
		$v_{21}^* = .983\text{cs}$ [9]
Benzene	$\rho_{38} = .859 \text{ gm/ml}$	$V_{40} = .503\text{cp}$ [8]
	$V_{38} = .52\text{cp}$	$V_{30} = .561\text{cp}$ [9]
	$v_{21} = .74\text{cs}$	$v_{21}^* = .742\text{cs}$ [8]
		$v_{21}^* = .737\text{cs}$ [9]
Ethylene glycol	$\rho_{38} = 1.101 \text{ gm/ml}$	$\rho_{30} = 1.106 \text{ gm/ml}$ [15]
	$V_{38} = 10.23\text{cp}$	$v_{25}^* \text{ approx.} = 15.6\text{cs}$ [9]
	$v_{21} = 15.8\text{cs}$	$v_{20}^* = 19.0\text{cs}$ [15]
Glycerol	$\rho_{38} = 1.250$	$v_{20}^* = 1182\text{cs}$ [8]
	$V_{38} = 320\text{cp}$	$v_{20}^* = 848\text{cs}$ [9]
	$v_{21} = 915\text{cs}$	
DC-710	$\rho_{38} = 1.089 \text{ gm/ml}$	$\rho_{25}^{**} = 1.107 \text{ gm/ml}$ [12]
	$V_{38} = 248\text{cp}$	$v_{25} = 500\text{cs}$ [12]
	$v_{21} = 566\text{cs}$	
FC-104	$\rho_{38} = 1.727 \text{ gm/ml}$	$\rho_{25} = 1.75 \text{ gm/ml}$ [11]
	$V_{38} = 1.02\text{cp}$	$v_{25} = .8\text{cs}$ [11]
	$v_{21} = .77\text{cs}$	

* Kinematic viscosity computed from reported absolute viscosity by: $v = V/\rho$. Density determined by multiplying specific gravity values [11] by density of water at reference temperature.

** In product literature specific gravity given at 25°C . Density determined by $\rho_{\text{DC-710}} = (\text{spec grav}_{25}) \times (\rho_{\text{H}_2\text{O}_{25}})$.

5. Considerations regarding n_2

In this paper we have considered liquids which may be applicable to high repetition rate Nd: glass lasers. Accordingly, one parameter of great interest is the nonlinear refractive index, n_2 , important for the propagation of high peak power laser pulses. The electronic part n_2^e of n_2 has been calculated according to the method of Boling, Glass and Owyong [5], and can be written

$$n_2^e (10^{-13} \text{esu}) = \frac{K(n_d - 1)(n_d^2 + 2)^2}{v_d \left\{ 1.517 + [(n_d^2 + 2)(n_d + 1)v_d] / 6n_d \right\}^{1/2}} \quad (5)$$

where K is an adjustable parameter (taken to be $K = 68$ here). The results for liquids in table 1 are given in table 3 and figure 2, where n_2^e has been plotted versus the linear refractive index n_d . As is the case for glasses, a low nonlinear index is generally correlated with a low linear index of refraction. In figure 1 we show n_d plotted versus v_d , and it is apparent that large Abbé values also correlate with liquids having low calculated values of n_2^e .

Although there is a paucity of data concerning experimentally determined n_2 values in liquids, we have been able to compare our calculated values of n_2^e for benzene with those found in the literature. The third order susceptibility $\chi_{1111}^{(3)}$ for a liquid or glass is related to the total n_2^T , through the relationship

$$n_2^T = \frac{12\pi}{n_d} \chi_{1111}^{(3)} \quad (6)$$

where

$$n_2^T = n_2^e + n_2^n \quad (7)$$

Here n_2^n is the nuclear contribution to n_2^T , and is usually in the range of 20-30% of n_2^T in glasses, but in liquids can be much larger. In this paper we concentrate strictly on n_2^e ; the nuclear contributions to n_2^T for liquids will be the subject of a separate publication. If we write $\chi_{1111}^{(3)}$ as a sum of electronic ($\chi_{1111}^{(3e)}$) and nuclear ($\chi_{1111}^{(3n)}$) parts

$$\chi_{1111}^{(3)} = \chi_{1111}^{(3e)} + \chi_{1111}^{(3n)} \quad (8)$$

then n_2^e is given by

$$n_2^e = \frac{12\pi}{n_d} \chi_{1111}^{(3e)} \quad (9)$$

Levenson and Bloembergen [16] have reported a value for $\chi_{1111}^{(3e)}$, as has Owyong [17]. Using eq. 9 to calculate n_2^e we arrive at the results shown in table 6 below.

Table 6. Calculated vs. experimental values for n_2^e in Benzene

$\frac{n_2^e (10^{-13} \text{esu})}{(\text{calculated})}$	$\frac{n_2^e (10^{-13} \text{esu})}{[16]}$	$\frac{n_2^e (10^{-13} \text{esu})}{[17]}$
3.1	4.62	5.02

Owyong [18] has determined that n_2^e is approximately 20% of n_2^T in benzene, and that $n_2^T = 14 \times 10^{-13} \text{esu}$; hence the nuclear part is $n_2^n = 10 \times 10^{-13} \text{esu}$. Our value for n_2^e is in fair agreement with both of the reported values, and will probably show better agreement once a data base has been established and the adjustable constant K in eq. 5 has been recalculated for liquids. Thus the data given in table 3 and figure 2 should be treated in a first order sense at present.

This is, to our knowledge, the first application of the Boling, Glass, Owyong theory [5] to the liquid state, and gives the electronic contribution n_2^e to the total nonlinear refractive index. Extension of this work to other liquids for which the total nonlinear refractive index is known will allow the separate contributions of electronic and nuclear susceptibility to be determined.

6. Discussion

Optimum characteristics for an index matching liquid or a coolant may be considered to be the following: low viscosity, high thermal conductivity, low absorption in important spectral regions, low dn/dT and low n_2 .

Low viscosity is an important property where liquid flows are considered. For example, glycerol exhibits relatively good values for n_2 , thermal conductivity and dn/dT which might lead one to consider glycerol as a coolant. However, the extremely large viscosity of glycerol would put a severe strain on any pumping system.

Heat removal considerations for cooling applications optimally require liquids with high thermal conductivities. A comparison of H_2O and FC-104 will serve as an example of this factor. Both liquids have low viscosities, low n_2 values, similar UV edges and FC-104 has lower absorption than H_2O , especially in the near-IR region. However, on the basis of thermal properties alone, H_2O would be the superior choice for a coolant as its thermal conductivity is 6 times greater than FC-104 and dn/dT for H_2O is approximately 1/3 lower. Another important thermal property is thermal diffusivity, defined as $K/(\rho C_p)$ where K = thermal conductivity, ρ = density, and C_p = specific heat at constant pressure. The specific heat of these liquids, not yet measured, is required to calculate this quantity. Another thermal measurement to be performed on these and other liquids at a later date is an examination of the coefficient of thermal expansion in the region around room temperature.

Characteristics other than those specifically mentioned above may come into play in an application decision. For example, silicone oils are quite difficult to dissolve with normal solvents. Hence, any cleanup of these materials can be a monumental task. Also silicones and TCP are examples of liquids which degrade in UV radiation. The UV edge of these liquids shifts towards the visible with exposure to intense UV from flashlamps. Care must also be used in choosing coolant/index matching liquids that are chemically compatible with materials used to contain them. $ZnCl_2$ (aq) is corrosive to aluminum and stainless steel, but does not react with 70/30 brass [19].

7. Conclusion

A wide range of values for refractive index, n_2 , dn/dT , thermal conductivity, density, viscosity, pH, and spectral transmittance is observed for the liquids examined in this work. The data generated in this investigation should aid in a system designer's decisions in choosing a particular liquid for a given application. Further investigation is planned of other useful properties including: thermal expansion, specific heat, thermal diffusivity, and $n_{1.15\mu m}$. This work will be expanded to consider additional liquids such as deuterated water, toluene, ethyl alcohol, cyclohexane, CCl_4 , and acetone.

8. Acknowledgements

This work was partially supported by the Laser Fusion Feasibility Project, which is sponsored by: Exxon Research and Engineering Company, General Electric Company, Northeast Utilities Company, Empire State Electric Energy Research Company, and New York State Energy Research and Development Administration.

9. References

- [1] N.I. Sax, Dangerous Properties of Industrial Materials, 4th ed. VanNostrand Reinhold Co., 1975.
- [2] J.V. Hughes, J. Sci. Inst., 18, 234 (1941).
- [3] Lafayette Instrument Co., Lafayette, Indiana.
- [4] R.W. Hellwarth, Phys. Rev., 152, 156 (1966).
- [5] N.L. Boling, A.J. Glass, A. Owyong, L³ report UCRL 75628 (1974).
- [6] B. Braun Melsungen A.G., San Mateo, Calif.
- [7] Sartorius-Werke GMBH, Göttingen, Germany.
- [8] R.C. Weast ed., Handbook of Chemistry and Physics, 52nd ed., Chemical Rubber Co., (1971).
- [9] J. Dean ed., Lange's Handbook of Chemistry, McGraw-Hill Book Co., 11th ed., 1973.
- [10] L. Meites, ed. Handbook of Analytical Chemistry, McGraw-Hill Book Co., 1st ed., 1963.
- [11] Product information on Fluorinert liquids, 3M Co., St. Paul, Minn.
- [12] Bulletin #22-281 (6/1974), Dow-Corning Corp., Midland, Mich.
- [13] D. Brown, S. Jacobs, J. Abate, O. Lewis, J. Rinefied, Laser Induced Damage in Optical Materials: 1977, NBS special pub., 509, 416.
- [14] Cargille Laboratories, Cedar Grove, NJ.
- [15] M. Windholz ed., The Merck Index, 9th ed., Merck & Co. Inc., 1977.
- [16] M.D. Levenson and N. Bloembergen, Phys. Rev. B, 10, 4447 (1974).
- [17] A. Owyong, Optics Comm., 16, 266 (1976).
- [18] A. Owyong, App. Phys. Lett. 26, 168 (1975).
- [19] LLE Annual Report 1977.

Figure 1. Abbe value versus refractive index.

Figure 2. Calculated nonlinear refractive index versus linear refractive index.

Figures 3-8

Spectral transmittance of liquids under study. All liquids scanned in 3.7 cm quartz cells at 50 Å/sec.

Figure 3.

Spectral transmittance of FC-104 and Benzene.

Figure 4. Spectral transmittance of H_2O and ZnCl_2 .

Figure 5. Spectral transmittance of ZnBr_2 and DC-710.

Figure 6. Spectral transmittance of Ethylene glycol and C5040.

Figure 7. Spectral transmittance of glycerol and C5610.

Figure 8. Spectral transmittance of Dimethyl sulfoxide and Tricresyl phosphate.

K. THERMAL CONDUCTIVITY @ 25°C cal/cm $^{\circ}$ C (10^{-3})

Figure 9. Thermal comparator signal versus thermal conductivity.