

LA-UR--88-414

DE88 006478

TITLE SOLID HOSTS FOR DYE LASER RODS - PART 1
CRITERIA FOR CHOOSING A HOST MATERIAL

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SUBMITTED TO LASERS 87 Meeting, Lake Tahoe, December 7-11, 1987
Proceedings

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SOLID HOSTS FOR DYE LASER RODS - PART 1
 CRITERIA FOR CHOOSING A HOST MATERIAL

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ABSTRACT

This paper will attempt to provide selection criteria for polymers as hosts for flashlamp-pumped dye laser rods. The properties of transparent polymer materials are compared with typical inorganic crystal and glass hosts

For some time now, our group, at the Los Alamos National Laboratory has been interested in trying to produce high quality inexpensive dye-doped laser rods. The potential low cost, in volume production of dye-doped plastic rods, combined with the wide variety of lasing wavelengths available from dyes was the primary driving force for this effort. The applications have been for both single and multiple shot operation, with fast flashlamp pumping. In the single shot case, if the dye-doped rod was damaged or destroyed after lasing once, this was acceptable as we were trying to get rather large power outputs from the rods. Since we needed simple, inexpensive, storable and compact systems, flowing liquid dye solvents were ruled out. This left us with the need to find suitable dye hosts that were solids with the necessary high optical quality. Organic polymers, namely plastics, such as the acrylics have been looked at for some time for these sorts of applications by other groups world wide.

As one gets more deeply involved with the problem of plastic dye laser hosts, a number of problems become apparent sooner or later. Aside from the need for very high optical quality in the plastic rods, one encounters problems with the nature of plastics as a class, when compared to inorganic crystal and glass laser hosts. Although dye-doped plastic materials may appear attractive at first glance from a potential low cost standpoint, particularly in volume production, a comparison of the physical properties of plastics versus inorganic hosts leads to some serious consequences. If one looks at the coefficient of thermal expansion and the thermal conductivity of plastics versus inorganic glasses and crystal hosts (Table 1), one sees that the coefficient of thermal expansion for plastics is, in general, one order of magnitude greater

TABLE 1.
 PROPERTIES OF PRINCIPAL OPTICAL PLASTICS

	Acrylics Polymethyl Methacrylate (Lucite) (Plexiglass)	Styrene Polystyrene (Dylene) (Styron) (Lucite)	MS Methyl Methacrylate Styrene Copolymer	SM Styrene Acrylonitrile (Lustran) (Tyl)	Polyacrylates (Lustran) (Marten)	TPX Methylpentene (TPX)	ABS ABS	ADC ADM Diphenyl Carbonate (CR 39)	Glass BK 7	
Refractive Index, n	Units									
n _d (589.3nm)		1.491	1.580	1.533-1.567	1.567-1.571	1.586	1.467	1.538	1.504	1.517
n _g (666.3nm)		1.488	1.588	1.558	1.563	1.581	1.464		1.501	1.512
n _f (688.1nm)		1.496	1.604	1.578	1.578	1.598	1.508		1.510	1.522
Abbe Value, V _d		61.4	31.1	38	37.8	34.5	11.4		56	64.6
Rate of Change in Index with Temperature	Δn/ΔT x 10 ³ /°C	8.8	12.0	14.0		11.8 to 14.3		14.3		+0.3
Coefficient of Linear Expansion	cm/cm x 10 ⁻⁶ /°C	6-7 @ 20°C	60-80		6.5-6.7	6-7.0		0.83	11.4	0.71
								25 to 75°C		
								14.6		
								15 to 125°C		
Distortion Temperature	°C	92	82		99-104	142		90		
1/4" from 2nd set	°C	101	110	100		146		84		
1/4" from 4th set	°C									
Recommended Max Core Temperature	°C	92	82	93	79-88	124			100	
Thermal Conductivity	cal/cm-cm ² -°C x 10 ⁻⁴	4.6	7.4-3.3	4.5	2.9	4.68	4.0		5	28.6
Moisture Absorption	%	2	3	3	3	3	5	17	3	
Luminescent Transmittance	% the area 3.175mm	92	88	90	88	89	90	79-90.6*	93	99.9
Water Absorption	% mm ² 24 hr @ 23°C	0.3	0.2	0.15	0.2-0.38	0.15			0.2	
Moisture Swellage	%	0.2-0.3	0.2-0.6	0.2	0.2-0.6	0.5-0.7	1.5-3.0			

* Luminescent Transmittance 75% Fluorescence 3.175mm 98.6% Fluorescence 3.175mm

for plastics than for the inorganic glasses and crystal hosts. The inverse is true for the thermal conductivity of plastics versus inorganic glasses. The thermal conductivity disparity of plastics, compared to many of the single crystal inorganic hosts such as sapphire and the garnets, is even more extreme because of the abnormally high thermal conductivity of these materials due to lattice phonon conduction. The change of refractive index of plastics as a function of temperature, dn/dt (Fig. 1), is closely related to the coefficient of thermal expansion. The combined effects of these two properties, as it affects plastic dye laser rods, is to aggravate the thermal stability problem by about the product of the dn/dt and the thermal conductivity ratios as compared to inorganic glasses or crystal hosts. The net result is that the optical thermal stability of plastic dye laser rods is about two orders of magnitude poorer than inorganic glass rods, approaching three orders of magnitude poorer than sapphire.

The overall effect of this very severe problem is that unless dye-doped plastic laser rods can be stored in an isothermal environment for a considerable length of time prior to firing the flashlamp, the optical properties of the plastic rod will be essentially useless if anything close to a few times diffraction limited output is needed. In the case of repetitively pulsed plastic dye laser rods, the thermal effects would be catastrophic.

A further problem related to the physical properties of plastic dye laser rods is the mechanical stability of the laser rod and cavity. Plastics are far less rigid or stiff than inorganic glass or crystal rods. Physical support under stress, vibration, G loads, etc. of plastic rods will be much more of a problem than conventional inorganic rods. Thermal gradients are apt to cause rod bending, which will throw the optical cavity out of alignment. Plastics are softer and scratch more easily than glass and most crystal hosts. About the only advantage the plastics have to offer, as far as physical properties are concerned, is resistance to mechanical and thermal shock and impact breakage. Being organic, plastics are also subject to photochemical damage. In addition, the laser dyes will undergo photochemical damage or bleaching. Photochemical damage of dye solutions is a problem in which the chemistry of the dye and the solvent system are synergistically inter-related. Polymerization without the use of initiators is desirable to obtain the best dye stability. For one shot applications, this is not too severe a problem if the rods do not bleach in long-term storage.

Probably more work has been done on acrylic, PMMA systems by numerous groups around the world than most other plastic systems for dye laser rod applications. Polymethyl methacrylate is by far the most common plastic in use for optics as it machines and polishes well, is easily moldable, has good environmental stability, and can have good uv transparency.

Acrylic's optical transmission is better than most optical plastics and can have low light scattering if clean and free of particulates (see Fig. 2). The optical damage threshold if clean is also quite good. The optical homogeneity of good quality molded acrylic optics is typically nearly two orders of magnitude poorer than good quality optical glass (see Table 1). Acrylic optics, even in well annealed, have considerable stress birefringence which is hard to remove. Adding the correct amount of a suitable plasticizer can correct the stress birefringence but can cause other problems such as loss of uv transparency, and poorer mechanical properties as well as possible interactions with the laser dye.

PMMA as such is a good solvent for the Coumarin family of dyes but is not a good solvent for Rhodamine or Kion Red dyes which require more polar solvents. Dye triplet state formation is often aggravated in plastic host solutions. Very recently, Dr. Rob Hermes of our Laboratory, has produced some extremely fine quality acrylic copolymer rods with good solvent properties, even for dyes requiring polar solvents.

In addition to PMMA, we have looked at a number of other polymer systems as candidates for plastic dye laser rods. We looked into some clear epoxies, a rigid polyurethane-polyester copolymer sold commercially by the name, Calthane 3200, and a number of organic glasses of the glycerol borate family. We also ordered some samples of some commercial, fluorescent dye loaded, extruded, PMMA rod from Bayer AG through their U.S. subsidiary, Mobay Corporation. This commercial, fluorescent, plastic rod and sheet is available in seven colors from deep red to violet and is very inexpensive.

The epoxies had little to commend them for our application as we had trouble with dye-hardener interactions, fairly poor ultra-violet transmission, poor optical quality, and poor solvent power for dyes requiring polar solvents. The Calthane 3200 looked somewhat more promising in that the uv transparency of this material is quite good. We never were able, however, to get good optical quality from this material and the solvent power for dyes needing polar solvents was again poor. The Bayer-Mobay commercial, fluorescent, PMMA extrusions had two problems. The material had both a fluorescent and a nonfluorescent dye dissolved in the host PMMA and the optical quality of the extrusions was very poor.

The family of organic glasses based on borate esters of polyhydric alcohols was rather interesting in that these materials could be produced with a hardness ranging from a brittle glass at room temperature (glycerol borate), to a high viscosity liquid at room temperature (propylene glycol borate). The third member of this group is ethylene glycol borate which is intermediate in hardness or viscosity between the brittle glycerol borate and the syrupy propylene glycol borate.

These three borate esters can be mixed together in any proportions to temper the hardness of the resultant organic glasses. We hoped that these organic glasses would be containable as very high viscosity semisolid glasses inside fused silica cylindrical dye cells which would behave like rigid dye rods. The severe T.C.E. mismatch between the high expansion organic glasses and the fused silica would be relieved by flow in the organic glasses. An expansion space for a bubble is needed to prevent cracking of the dye cell when subjected to temperature changes. The viscosity of the organic glass is so high as to prevent convection from disturbing the optical quality of the laser medium. The glycol borate esters are again poor solvents for dyes requiring polar solvents but their uv transmission is quite good. This exercise was not successful for a number of reasons. The most severe problem is that these glycol borate esters seem to produce a swarm of tiny gas bubbles when subjected to the intense light of the xenon flashlamp and these bubbles can't be removed from the optical path without remelting the organic glass to reduce the viscosity to a low value.

One other possibility came to mind while we were working on plastic laser rods. The severe problems with thermally induced optical inhomogeneities in any high T.C.E. and low thermal conductivity, plastic dye laser rod might possibly be sidestepped by making a rigid, bonded, fiberoptic bundle, laser rod. This rod would have a dye doped, higher refractive index, plastic core material, surrounded by a clear, undoped, lower index, plastic cladding which constitutes a very thin coating between the adjacent dye doped core fibers. The solid matrix of cladding would couple the pump light into the matrix and the individual dye doped fibers would be mode coupled to one another by the evanescent wave penetrating the thin cladding between adjacent fibers. This would tend to prevent all the individual fibers in the bundle from lasing unsynchronized with one another which would make the entire fiberoptic laser bundle act as a more or less incoherent light source. If the evanescent wave coupling between adjacent fibers in the matrix works, the optical coherence should be greatly improved. The partitioning of the laser rod into many small lasing regions, each isolated from its neighbor, except for the evanescent wave coupling, would permit fairly large refractive index gradients across the cladding to exist without disturbing the lasing medium, which is the individual high index dye doped fibers. Since each fiber is bounded by the steep index gradient at the core cladding interface, the laser light would be trapped and confined to the fiber it originated in. The laser cavity mirrors would have to be in contact with the cut and polished ends of the laser fiberoptic bundle. Transverse pump light should be well coupled throughout the bundle. There would be an effective change in the cavity length across the width of the fiberoptic bundle when subjected to a temperature gradient, however, to what extent these different coupled lasers can synchronize with slightly different cavity lengths is unknown at this time.

Fabrication of such a structure although complicated might not be as difficult to accomplish at a reasonable cost as it may seem at first glance. Individual dye-doped plastic fiber with a higher refractive index than the clear cladding material could be produced in long lengths. The thin layer of clear cladding could be applied as a dip coating and cured or dried by solvent evaporation. This thinly clad fiber could then be bundled and pressure bonded, perhaps by warm isostatic pressing or warm extrusion to form the laser fiberoptic bundle. This material could then be cut to length, polished and have the flat-flat cavity mirrors cemented on the ends. The cavity mirrors may be able to be simply deposited on the polished ends of the bundle by physical vapor deposition methods and further reduce the cost of the fiberoptic laser rods. If this scheme works, it pretty much sidesteps most of the thermal problems associated with ordinary plastic dye laser rods. This system could possibly operate as a ring laser since a curved optical path could be made in a fiberoptic bundle. This device might have some interesting possibilities.

Table 1, Figures 1 and 2: "Optical Plastics: Properties and Tolerances," The Photonics Design and Applications Handbook, Book 2, 33rd Edition, pp H225-H230 (1987).

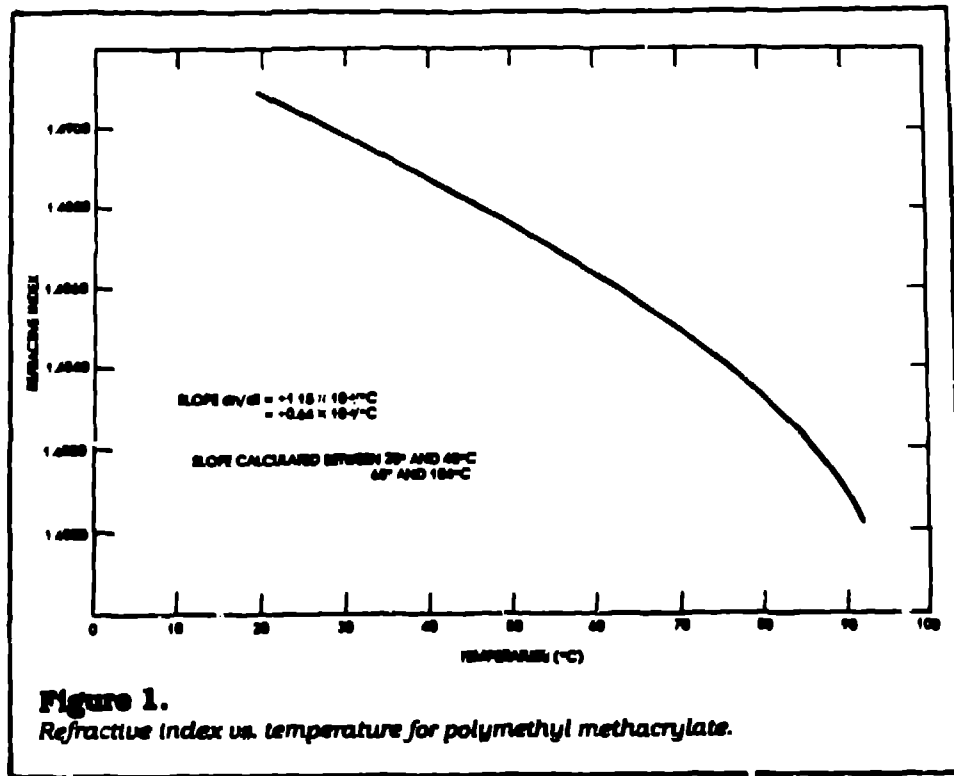


Figure 1.
 Refractive index vs. temperature for polymethyl methacrylate.

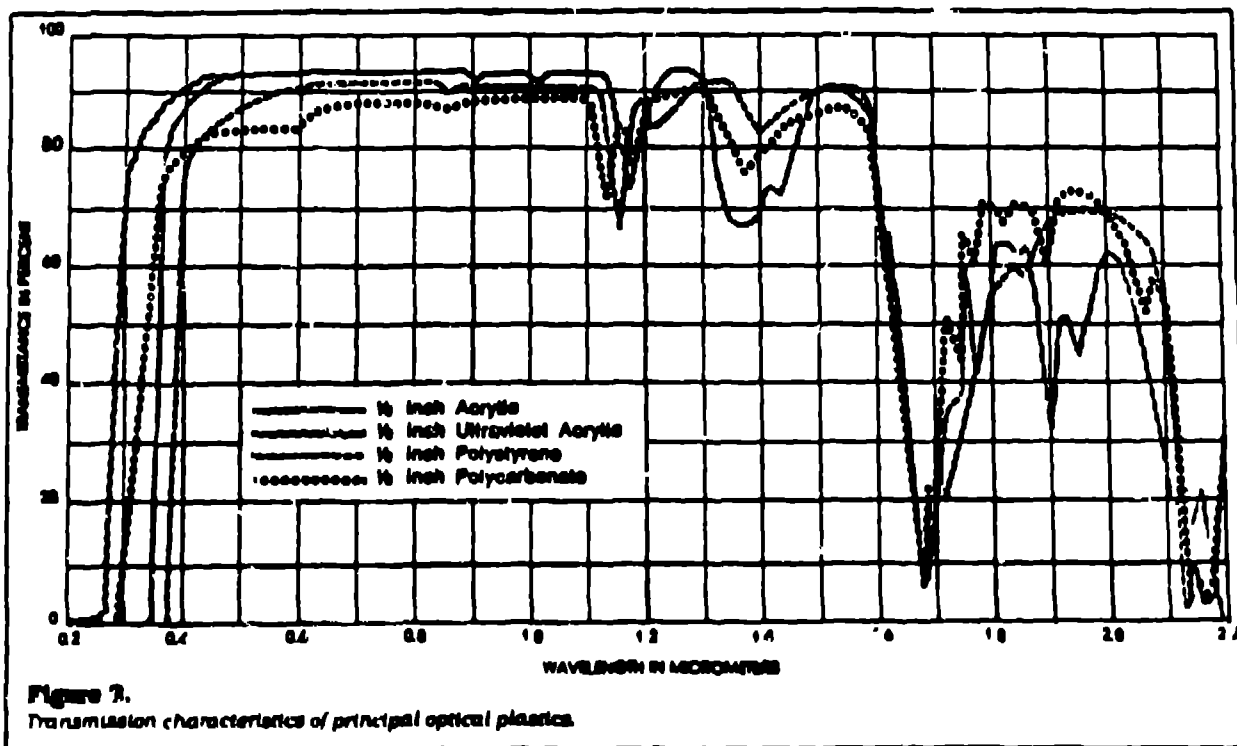


Figure 2.
 Transmission characteristics of principal optical plastics.