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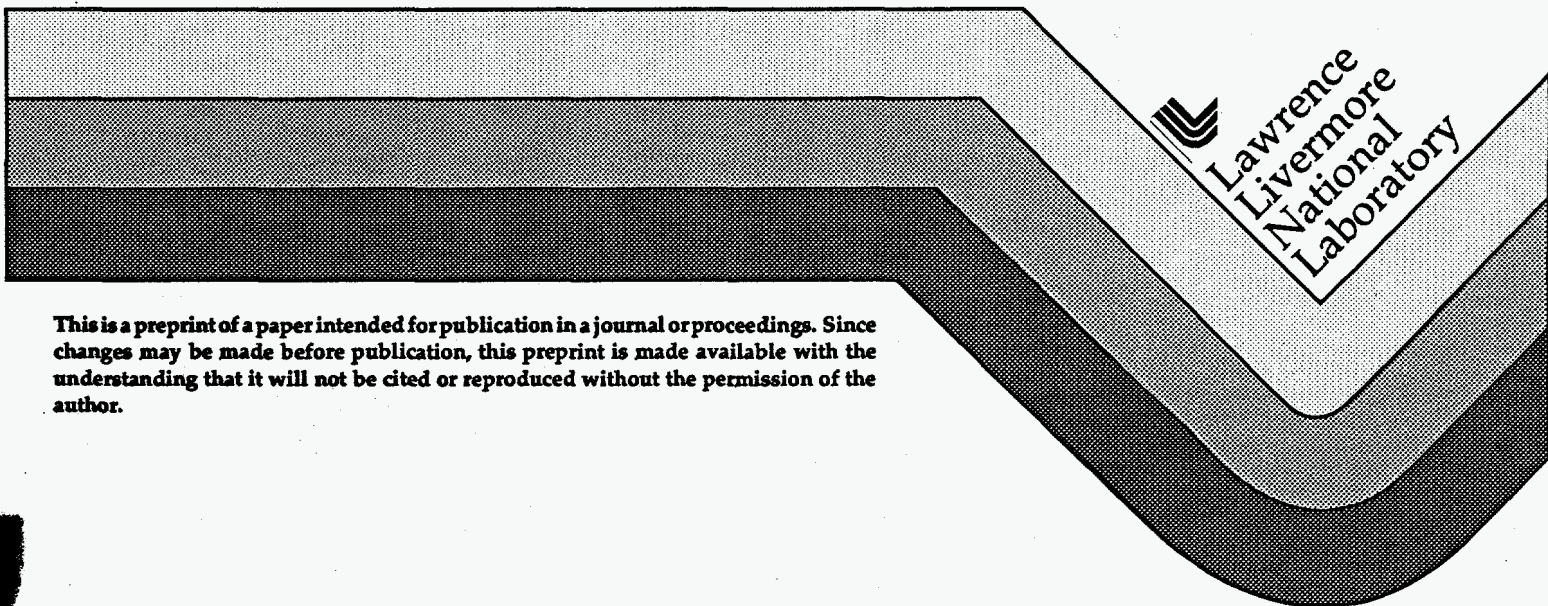
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PREPRINT

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Properties of a New Average Power Nd-Doped Phosphate Glass

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ABSTRACT

The Nd-doped phosphate laser glass described herein can withstand 2.3 times greater thermal loading without fracture, compared to APG-1 (commercially-available average-power glass from Schott Glass Technologies). The enhanced thermal loading capability is established on the basis of the intrinsic thermomechanical properties and by direct thermally-induced fracture experiments using Ar-ion laser heating of the samples. This Nd-doped phosphate glass (referred to as APG-t) is found to be characterized by a 29% lower gain cross section and a 25% longer low-concentration emission lifetime.

1. INTRODUCTION

Nd-doped glasses have been deployed for 1.05 μm laser systems for several decades now, and scientists have come to understand how the properties of these glasses impact the laser performance. Over the years, Nd:glass-based systems have tended to involve lasers dedicated to generating high single-shot energies, high average power, or ultrashort pulses. All three of these areas have continued to experience intense interest to the present day.

An important step in laser glasses occurred in 1967 when phosphate-based compositions were first explored [1]. Phosphates were found to have several important attributes over silicate glasses [2]. In particular, Nd:phosphate glasses provide greater extraction efficiency [3] (by virtue of reduced hole-burning and a higher gain cross section), and can also be manufactured free of platinum inclusions [4] (thereby greatly increasing the optical damage threshold). Since micron-size platinum inclusions appear to be an inevitable consequence of today's glass melting technology, it

is of crucial significance to note that it is possible to chemically treat phosphate glasses so as to completely eliminate these particles. It has not yet been possible to achieve comparable results for silicates or fluorides.

If we proceed with the assumption that phosphates are the preferred medium for most (non-fiber) Nd:glass laser applications, then one is compelled to evaluate the range of properties available from this class of materials. In this article we discuss our research on the development of phosphate glasses that offer an enhanced thermal fracture limit. Among the laser glasses in common usage are LG-750 and APG-1 (from Schott Glass Technologies), in addition to Nd:phosphate glasses produced by Hoya [5], Kigre [6] and other companies. The result of our campaign to define a Nd:glass with improved thermomechanical properties is the so-called APG-t glass, where "t" refers to "test-glass" since this glass is not yet commercially available. We describe the properties of APG-t in the context of the established characteristics of LG-750 (fusion laser glass) and APG-1 (average-power glass), in order to emphasize the uniqueness of the new glass discussed herein.

2. OPTICAL PROPERTIES

The absorption spectrum appears in Fig. 1 where it is plotted on an absolute scale by correcting the optical density for the Nd concentration and sample length. The absorption spectrum is similar to that of many other Nd-doped phosphate glasses. The result of exciting the sample with a Xe lamp and monitoring the emission is displayed in Fig. 2 (after being corrected for the spectral response function). The emission width, defined as

$$\Delta\lambda_{em} = \frac{\int I_{em}(\lambda) d\lambda}{I_{em}(\lambda_p)}, \quad (1)$$

is found to be $\Delta\lambda_{em} = 31.5$ nm for APG-t, where $\lambda_p = 1054.6$ nm. The peak emission cross section may be calculated with the Einstein relation [7],

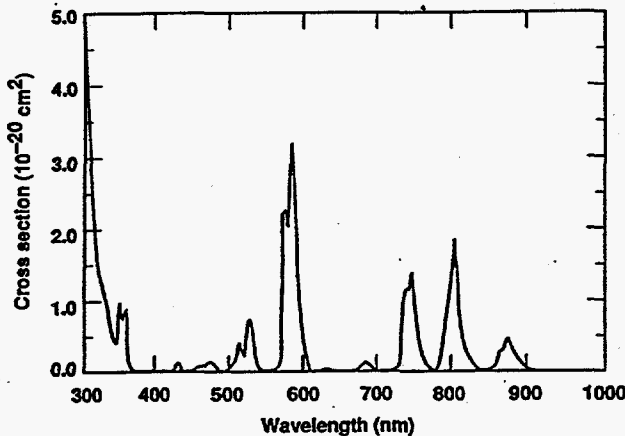


Figure 1. Absorption spectrum of APG-t glass plotted versus wavelength on an absolute cross section scale.

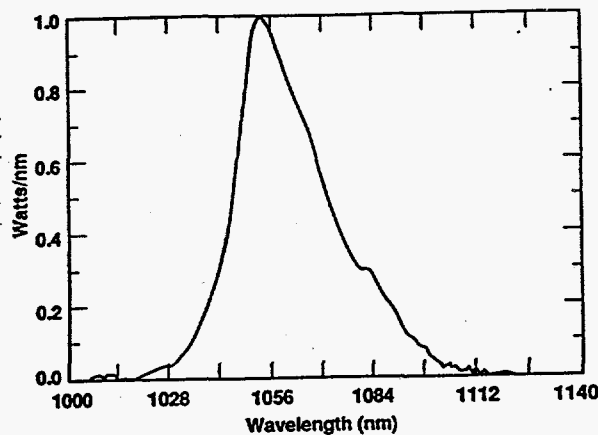


Figure 2. Emission spectrum of APG-t arising from the ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition.

where the radiative lifetime τ_{rad} and branching ratio $\beta_{11/2}$ are required.

One way to determine $\beta_{11/2}$ and τ_{rad} is to use the theory of Judd and Ofelt (JO) [8]. For the case of APG-t they are (in units of 10^{-20} cm^2): $\Omega_2 = 5.60$, $\Omega_4 = 4.02$, and $\Omega_6 = 4.22$, yielding $\tau_{rad}(\text{JO}) = 451 \pm 20$ μsec . By similar means we can use the JO theory to determine that the fractional

emission into the ${}^4F_{3/2} \rightarrow {}^4I_{11/2}$ transition (compared to all the 4I_j) is $\beta_{11/2} = 0.480$.

Since $\tau_{rad}(\text{JO})$ is derived solely from the absorption spectra, we also evaluate the radiative lifetime by a second, more direct method. In these experiments [9], we obtained the emission lifetime (τ_{em}) as well as a measurement of the quantum yield (η_{QY}) in order to provide an independent assessment of the radiative lifetime using the equation: $\tau_{rad}(\text{QY}) = \tau_{em}/\eta_{QY}$. The radiative lifetime deduced by this method is $\tau_{rad}(\text{QY}) = 455 \pm 30$ μsec . Accordingly, we employed the average of the JO and QY values, or $\tau_{rad} = 453$ μsec , to find that the peak emission cross section is $\sigma_{em}(\lambda_p) = 2.39 \times 10^{-20} \text{ cm}^2$; the cross sections of LG-750 and APG-1 are 3.68 and 3.35, respectively.

A well-known issue impacting all Nd-doped materials is that of concentration quenching [18]. The measured lifetimes τ_{meas} had to be corrected for two effects, including radiation trapping and water quenching [9,10] to yield the true emission lifetime, τ_{em} . We plot τ_{em} versus the Nd concentration N_{Nd} in units of 10^{20} cm^{-3} , as shown in Fig. 3. Here, similar data acquired for the commercial laser glasses APG-1 and LG-750 are included for comparisons; the radiative lifetimes (see above) are plotted to describe the zero concentration limit. The APG-t data points of Fig. 3 are observed to fall on a simple straight line.

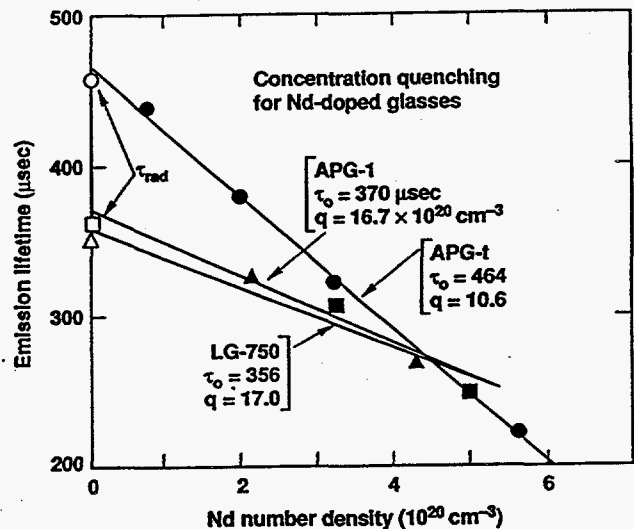


Figure 3. Plot of the Nd^{3+} emission lifetime as a function of dopant concentration for APG-t, APG-1 and LG-750.

In lieu of analyzing the data on the basis of theories requiring fitting to several parameters [10], we employ an empirical linear approach with

$$\tau_{em} = \tau_0 (1 - N_{Nd}/q) \quad (2)$$

for $N_{Nd} \leq q/2$, and obtain $\tau_0 = 464 \mu\text{sec}$ and $q = 10.6 \times 10^{20} \text{cm}^{-3}$.

3. LASER ACTION

As an additional exercise in assessing the viability of the APG-t laser glass, we constructed a simple laser cavity to demonstrate that the material exhibited laser action [11]. In these experiments, we used a 10 cm long nearly concentric cavity and a 514 nm Ar ion laser pump source. Since thermal lensing began to degrade the laser performance for a duty cycle of $>10\%$, a 2% duty cycle was utilized to eliminate any concerns of this nature. Figure 4 contains plots of the laser output power as a function of the absorbed input power (expressed as instantaneous rather than average powers). As we see from the data the average slope efficiency is 13.9% for 3.1% output coupling, and 21.9% for a 6.3% output coupler. The observed increase in slope efficiency η with greater output coupling T , is often indicative of the presence of passive loss in the cavity. The impact of the double-pass loss L present in the laser glass is described with

$$\eta = \eta_0 \frac{T}{T+L} \quad (3)$$

where η_0 is the so-called intrinsic efficiency. From the laser data in Fig. 4 we calculate that $L = 8.3\%$ and $\eta_0 = 51\%$. In the absence of dynamic losses (e.g., excited state absorption), the intrinsic efficiency is given by the ratio of the pump and laser wavelengths or the quantum defect, $\eta_0 = \lambda_p/\lambda_\ell = 49\%$. We therefore find that the measured and calculated magnitudes of η_0 agree. The double-pass loss of 8.3% may be converted into a loss coefficient of 6.4%/cm.

We are uncertain as to the origin of the loss at this time, in that the measured scatter losses at the laser wavelength are $<0.1\%$ and the absorption losses at the laser wavelength are 1.2%/cm. Remaining possibilities could entail the impact of wavefront distortion introduced by the sample, incomplete compensation for the reflectivity of the sample surfaces, or losses related to stress-induced birefringence.

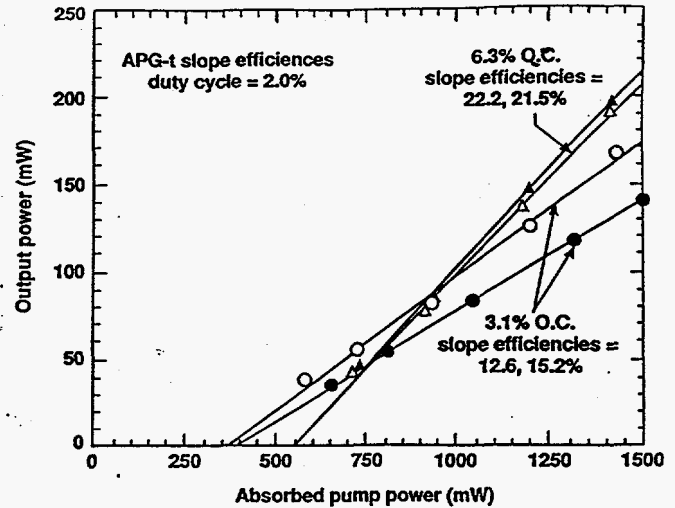


Figure 4. Laser performance of APG-t obtained for two values of output coupling (o.c.), using a 514 nm pump source and a 10 cm nearly-concentric cavity.

4. THERMAL AND MECHANICAL PROPERTIES

The ability of a phosphate laser glass to sustain internal thermal gradients without fracture is an important characteristic for high average power applications. The primary predicted advantage of APG-t over other phosphate glasses is its superior thermal and mechanical properties. In an effort to experimentally establish the expected superior thermal properties of APG-t, an experiment was devised that would demonstrate the fracture probability in the presence of thermal gradients. Nd-doped APG-t, APG-1 and LG-750 laser glasses containing 3.5%, 3.43%, and 4.0% by weight, respectively, were fabricated into $\sim 3 \times 3 \times 3$ mm cubes (~ 10 for each glass type) with laser quality polishes on two opposing sides. These glass samples were then irradiated with 514 nm light from an argon-ion laser. The argon-ion beam was variably attenuated (0 to 5W) and focused with a 7.5 cm focal length lens to a 40 μm diameter ($1/e$) near-Gaussian spot size. The samples were tested by first starting out at relatively low 514 nm power levels and then translating each sample back and forth across one transverse dimension and observing whether catastrophic cracking occurred. The beam power was then increased in small incremental steps with similar sample translations being performed between each step. When fracture was observed, the incident power was recorded. This procedure was repeated for each of

the samples. The deposited thermal power density, P_{th}/V , was calculated by including the optical absorption fraction, surface reflection, and the fraction of optical power that is nonradiative (i.e., leading to heating). The fracture probability was determined by ranking the glasses in order of the thermal power density at which they fractured and assigning a fracture probability via the relation [12,13]

$$\Phi = (n - 0.5)/N \quad (4)$$

where n is the rank and N is the total number of samples of each glass type. The data appears in this format in Fig. 5, where the solid lines are numerical fits using Weibull statistics that will be discussed in more detail later in this section.

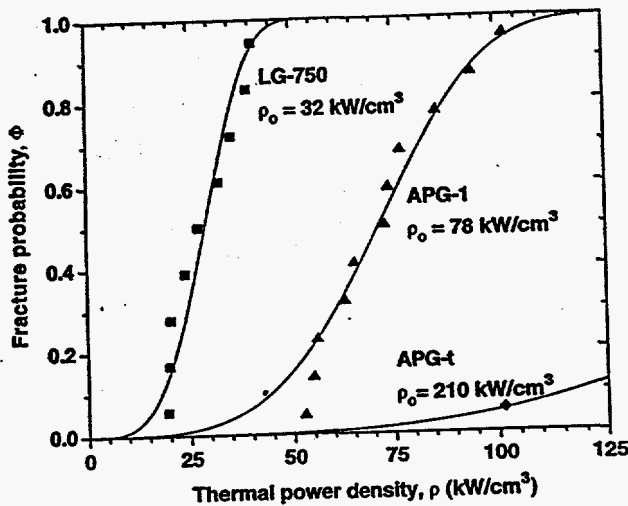


Figure 5. Thermal fracture results for APG-t, APG-1, and LG-750 glasses. The solid lines are numerical fits to the experimental data points using Weibull statistics to describe the fracture probability.

The data clearly shows that LG-750 tends to fracture at lower thermal power density than is the case for APG-1, as one would expect from previous thermal-mechanical studies [14], while APG-t exhibits a lower thermal fracture probability than APG-1 for a given thermal power density. Although only one sample of APG-t was observed to crack under thermal loading, there were eight other samples that were subjected to >100 kW/cm³ for which no fracture was observed.

Weibull statistics can be employed to describe thermally induced fracture for samples with a given distribution of surface flaw sizes determined

primarily by the surface polish technique. This treatment assumes that the sample cracks when the weakest point (corresponding to the location with the largest flaw) of the sample fails. The fracture probability for a two parameter Weibull distribution can then be written as [15,16]

$$\Phi = 1 - \exp\left[-\left(\frac{\rho}{\rho_0}\right)^m\right], \quad (5)$$

where m and ρ_0 are the adjustable shape and scaling parameters and ρ is the thermal power density expressed in units of power per unit volume. A least squares fitting routine was utilized for each of the three glass types to relate the data in Fig. 5 to the two adjustable parameters in Eq. 5. The scaling parameter, ρ_0 , was varied independently for each sample since this parameter is expected to be proportional to the intrinsic ability of the glass to withstand fracturing. The shape parameter, m , was held constant between samples since this parameter is related to the distribution of flaw sizes left behind after polishing and all of the sample surfaces were prepared with the same polishing technique. The sum of the least squares errors of the three glass types was minimized. A value of $m = 4.0$ was found to simultaneously provide the best fit to the shape of the APG-1 and LG-750 data sets. The scaling parameters, ρ_0 , then become 31.7 ± 1.3 , 78.4 ± 0.9 , and 210 ± 50 kW/cm³ for LG-750, APG-1 and APG-t, respectively.

The question naturally arises as to whether the scaling parameter, ρ_0 , is proportional to what one would expect from theoretical predictions of the thermal shock parameter R_T , calculated from other measured material properties. The thermal shock parameter serves as a thermal figure of merit for high average power applications, and is directly related to the maximum thermal load which a surface cooled laser slab can withstand without fracture. It is expressed as [13]

$$R_T = \frac{K_{Ic} \kappa (1 - \nu)}{\alpha_{exp} E \sqrt{a}} \quad (6)$$

where K_{Ic} , κ , ν , α_{exp} , E , and a are the fracture toughness, thermal conductivity, Poisson's ratio, thermal expansion coefficient, Young's modulus, and the radius of the damage initiating flaw size, respectively. By assuming a flaw size of $2a = 50$ μ m which is typical for mechanically polished surfaces [13], one obtains thermal shock parameters of 255, 111, and 46 W/m for APG-t, APG-1 and LG-750, respectively [14]. It is readily apparent that the thermal shock parameter (R_T) is proportional to

the fracture scaling parameter (ρ_0) to within the error bars. Consequently, we believe that the above fracture experiments, which indicate that APG-t is significantly more fracture resistant than currently available high average power phosphate glasses such as APG-1, are validated by agreement with thermal shock parameter calculations. In this way, our somewhat tenuous fit through a single point in Fig. 5 has been validated on the basis of the inherent thermomechanical properties of the glasses.

5. SUMMARY

APG-t appears to be a viable Nd-doped phosphate glass that offers a new combination of laser, optical, thermal, and mechanical properties. In comparing APG-t to two other glasses that are commercially available from Schott Glass Technologies (LG-750 and APG-1), it becomes apparent that various trade-offs among the material properties are possible. For example, APG-t has a lower emission cross section σ_{em} , although the thermal shock parameter R_T is substantially enhanced. Similarly, the concentration quenching is steeper for APG-t while the zero-concentration lifetime is longer (permitting greater energy storage for lightly-doped samples). We lastly note in passing that the thermal lensing of APG-t is expected to be ~40% stronger than is the case for APG-1.

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