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# **PTR GLASS MELTING FACILITY FOR FABRICATION OF DIFFRACTIVE OPTICAL ELEMENTS**

(Final report)

Acc. # 6501-6062, DURIP MDA Contract F49620-02-1-0303.

May 15, 2004

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### **Abstract**

The proposed research instrumentation, a PTR Glass Melting Facility, is used to support MDA-funded and new research initiatives in the study of photosensitive glass for rugged volume diffractive optical elements. This instrumentation significantly impacts our existing effort in hologram recording in photo-thermo-refractive (PTR) glasses. Diffractive optical elements made from this glass are in the BMDO/MDA funded program "Holographic Narrow-Band Filter and Data Storage for Optical Target Characterization" and a number of PTR related projects funded by DARPA, AFRL, and ARL. Our significant achievements in the creation of the new type of diffractive optical elements were enabled by a "COMPUTER CONTROLLED RECORDING AND TESTING SYSTEM FOR STUDIES OF HOLOGRAPHIC OPTICAL ELEMENTS" that was designed and constructed under BMDO DURIP project that was started in year 2000. The level of current and future efforts limited by having to use 400 ml fused silica crucibles for melting PTR glass. The new melting system is based on the use of a platinum crucible that allows fabricating PTR glass blanks with diameter of up to 50 mm having a homogeneity of refractive index of better than  $10^{-5}$ . This homogeneous photosensitive glass enables creation of diffractive optical components with ultimate spectral and angular selectivity that are necessary for the further development of the PTR technology in different application for high power laser beam control and optical target characterization. Glass produced by the requested system will be utilized in new research efforts for high-accuracy holographic optical elements is used in laser and opto-electronic devices for optical target characterization, such as narrow band filters, attenuators, beam splitters, samplers, multiplexers, etc.

### **1. Introduction: PTR glass – a new recording medium for phase holograms**

One of the new approaches to the optical design of laser systems is the use of diffractive optical components including such very promising elements as volume diffractive (Bragg) gratings.

However, their application is restrained by the lack of available photosensitive materials for volume hologram recording [1]. Existing holographic materials are silver halide photographic emulsions, dichromated gelatin, photoresists, photopolymers, photothermoplastics, polymers with spectral hole-burning, and photorefractive crystals. Organic materials (photographic emulsions, dichromated gelatin, and photopolymers) are sensitive to humidity. Moreover, they significantly shrink in the development process. Inorganic materials (photorefractive crystals) have low resistance to elevated temperatures and produce additional patterns because of exposure to the beam diffracted from the recorded grating. It is important to note that all these materials have significant losses (absorption and scattering) at laser wavelengths and low tolerance to laser radiation.

The other approach to volume hologram recording in inorganic glass was realized in Ref. [2 – 4] in lithium-aluminum-silicate [2] and sodium-zinc-aluminum-silicate [3-4] glasses doped with silver and cerium, by exposure to UV radiation followed by thermal treatment. Holograms in these glasses had relative diffraction efficiency up to 90 % and transmission up to 50 %, were stable up to 400°C. High diffraction efficiency means that phase pattern (spatial modulation of refractive index) was recorded. This phenomenon was named the “photo-thermo-refractive” process. Glasses, which possess such properties, were named “photo-thermo-refractive” (PTR) glasses. Recent efforts in improvement of glass fabrication, exposure, and thermal development [5-8] resulted in absolute diffraction efficiency of holograms in PTR glasses increase above 95 %. This technology of PTR glass and robust diffractive optical elements was protected by two US patents [9, 10]. Thus, this material can be classified as a new promising medium for three-dimensional hologram recording.

The photo-thermal process is based on precipitation of dielectric microcrystals in the bulk of glass exposed to UV radiation (Fig. 1). The first step is the exposure of the glass sample to the near UV radiation, which produces an ionization of a cerium ion. The electrons released from the cerium are then trapped by a silver ion

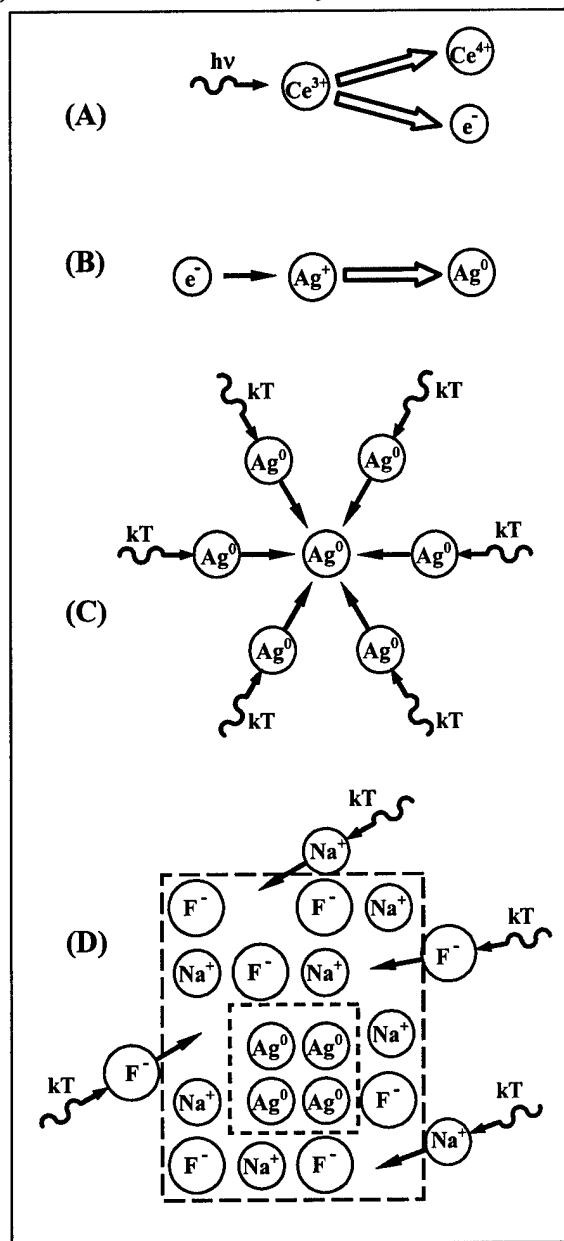


Fig. 1. Stages of the photo-thermo-refractive process. A - photoionization of  $Ce^{3+}$ , B - photoreduction of  $Ag^+$ , C - thermal aggregation of  $Ag^0$ , D - thermal crystallization of NaF. A and B occurs at room temperature, C - at 450°C, D- at 520°C

converting it to a neutral atom. This second stage corresponds to a latent image formation in conventional photo-materials and no significant coloration or refractive index variations occur in PTR glass at this stage. The third stage is the diffusion of silver atoms, which leads to creation of tiny silver crystals at temperatures 450-500°C. These silver particles serve as the nucleation centers for sodium fluoride crystal growth at temperatures between 500°C and 550°C. It was found that after this stage, a refractive index of exposed area decreases for about  $10^{-3}$ .

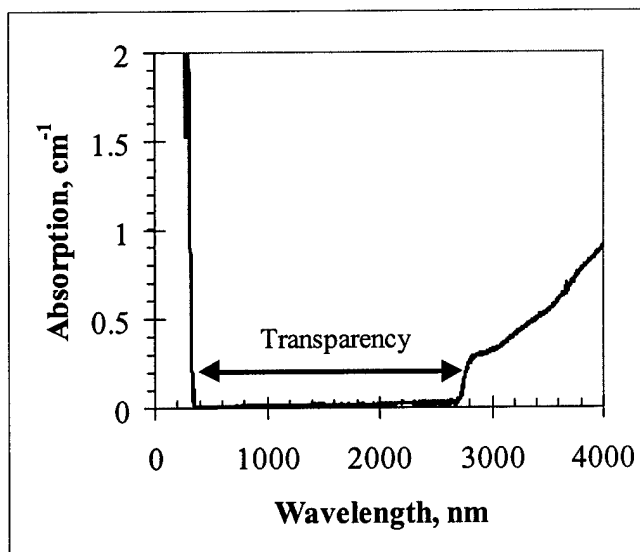


Fig. 2. Absorption spectrum of PTR glass

Absorption spectrum of PTR glass is presented in Fig. 2. The window of complete transparency is from 350 to 2700 nm. Absorption edge in the UV region is produced by admixtures of silver and cerium contained in PTR glass. Absorption at wavelengths greater  $2.7 \mu\text{m}$  is produced by hydroxyl groups contained in each silicate glass as technological contaminations. An additional absorption of PTR glass undergoing UV exposure at 325 nm is in the UV region. Thermal development causes an additional absorption in the visible region. Fluoride crystals are colorless and therefore cannot cause an additional absorption but they can cause an induced scattering. Additional absorption does not exceed  $0.3 \text{ cm}^{-1}$  and  $0.03 \text{ cm}^{-1}$  in the blue and red regions, respectively. Additional absorption in the whole IR region is not detectable by spectrophotometer.

Fig. 3 shows the UV part of absorption spectrum of a virgin PTR glass with an absorption band of  $\text{Ce}^{3+}$  having maximum at 305 nm. Excitation of PTR glass within this absorption band produces free electron and converts an absorbing ion to  $\text{Ce}^{4+}$  form. The released electron triggers the whole chain of photo-thermo-induced structural transformations in glass. The short wavelength edge, at which writing radiation is attenuated by two times in the recording medium (an optical density is about 0.3) is placed at 330 nm for a 1 cm thick plate and at 265 nm for 1 mm thick plate. This means that the use of UV lasers with wavelengths greater 300 nm allows writing of uniform gratings with thickness exceeding 1 mm.

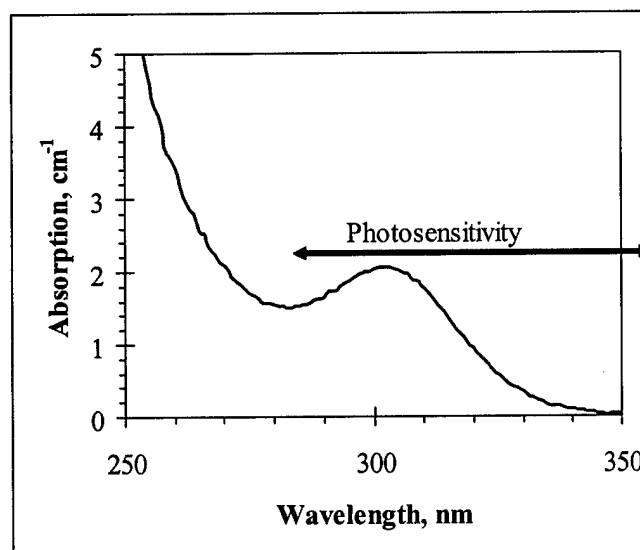


Fig. 3. UV absorption spectrum of PTR glass

Dependence of the induced index of refraction on dosage for different times of thermal development is shown in Fig. 4. One can see that refractive index decreases for about  $6 \times 10^{-4}$  (600 ppm) after exposure and thermal development. An induced refractive index exhibited a curve with a continuously decreasing slope. One can see that increasing of the time of development increases the initial slope of the dependencies which means increasing of the actual photosensitivity. However, the further development leads to the saturation of the induced index of refraction. This result allows the choice of optimal regime for the development. It was found that there is no difference in the refractive index decrement for the same dosage received for different values of irradiance. This means that PTR glass obeys the Reciprocity Law and the induced index of refraction depends on the total dosage only. The maximum value of the refractive index decrement in our experiments was about 1000 ppm. The refractive index decrement depends linearly on dosage up to 300 ppm and shows saturation at higher values. An approximate value of the photosensitivity (dosage derivative of a refractive index increment) of PTR glass melted in silica crucible for 325 nm irradiation followed by 3 hours of development at 520°C is  $1.5 \text{ ppm} \cdot \text{cm}^2/\text{mJ}$ .

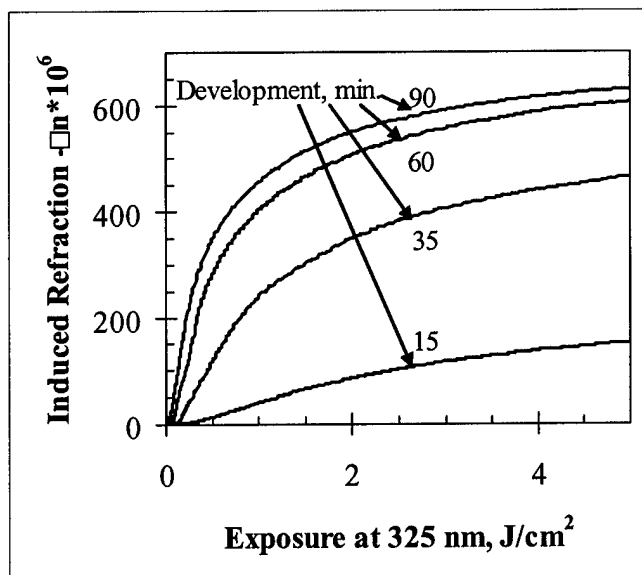


Fig. 4. Induced index of refraction in PTR glass after exposure to UV radiation at 325 nm and thermal development at 520°C

Bragg gratings in PTR glass were recorded by an exposure to the interference pattern of radiation of a He-Cd laser operating at 325 nm. Spatial frequency was varied from  $50 \text{ mm}^{-1}$  up to about  $10,000 \text{ mm}^{-1}$ . Volume gratings in both transmitting and reflecting mode were recorded with thickness ranged from 0.5 mm to 10 mm. Diffraction efficiency was measured at 633 nm, 1064 nm, and 1550 nm. In all cases, an absolute diffraction efficiency of PTR Bragg gratings exceeds 95 % of the theoretical value. The angular and spectral selectivity of both transmitting and reflecting PTR gratings did not exceed the theoretical value for more than 10 %. Minimum values of spectral and angular selectivity for PTR Bragg grating having thickness below 3 mm were in the range of tens of picometers and hundreds of microradians, respectively. It was shown that hologram aging at temperatures up to 400°C does not reduce diffraction efficiency. An expose of developed holograms to visible and UV radiation does not cause hologram destruction. Laser damage threshold of PTR glasses in nanosecond region is about  $40 \text{ J/cm}^2$  for 8 ns laser pulses [11]. Recent testing [8] has shown that PTR Bragg grating survived 30 min in  $100 \text{ kW/cm}^2$  laser beam at 1080 nm with no evidence of destruction or fading.

Thus, PTR Bragg gratings are the unique diffractive components which are finding the wide application in a number of laser system designs in such DoD funded projects as "Holographic narrow-band filter and data storage for optical target characterization," "High efficient

holographic narrow-band mirror for pulsed neodymium laser,” “Bragg selectors of transversal modes for pulsed neodymium laser,” and “Holographic filters with adjustable transmittance for high power lasers” for MDA, “Angularly addressed holographic optical elements in PTR glasses for 1.5  $\mu$ ” for STAB program at DARPA, “Improved technology for larger aperture Bragg deflectors in PTR glass” for THOR program at DARPA, “Incoherent combining of high-power laser beams by volume diffractive gratings in a photosensitive silicate glass” for AFOSR, “Volume diffractive optical elements for high-power laser beam control” for ARL, “Photo-thermo-refractive Bragg mirrors for high efficiency diode sources” for DARPA.

## 2. Technology of PTR glass

The photo-thermo-refractive glass is a sodium-zinc-aluminum-silicate glass doped with cerium, silver, bromine, and fluorine. There are three main problems in the PTR glass technology. The first one is the traditional problem for all optical glasses – optical homogeneity. This means that a refractive index of the glass casting (or boule) should be the same in all its parts. The usual requirements for the tolerable spatial fluctuations of refractive index are ranged from  $10^{-4}$  (cheap lenses) to  $10^{-6}$  in space objectives. In the case of the diffractive optical elements this requirement is about of  $10^{-5}$ . The conventional technology of the homogeneous optical glasses is based on melting and stirring of the melt in the large crucibles (usually they are of tens or hundreds of liters). We had no such opportunity to develop so expensive technology at the university and could not offer it to the start-up company at the stage of research and development. Therefore we had to develop a technology for homogeneous glass fabrication in small crucibles.

The second problem is the presence of such volatile components as bromine and fluorine in the PTR glass composition compare to the usual oxide optical glass composition. Traditional technology of fluorine containing optical glass uses melting in an isolated volume produced by special construction of platinum crucible and stirrer including the hydro-seal. We will consider such an approach for the next section but we had to avoid it at the stage of research and therefore we had to find a way to obtain homogeneous areas of fluorine containing glass fabricated in the open silica crucibles.

The third problem is caused by an additional requirement of homogeneity of photosensitivity imposed by the use of PTR glass not only as an optical glass but as a photosensitive medium for the volume hologram recording.

To solve the mentioned problems, the following technology was developed. The photosensitive glass of approximate

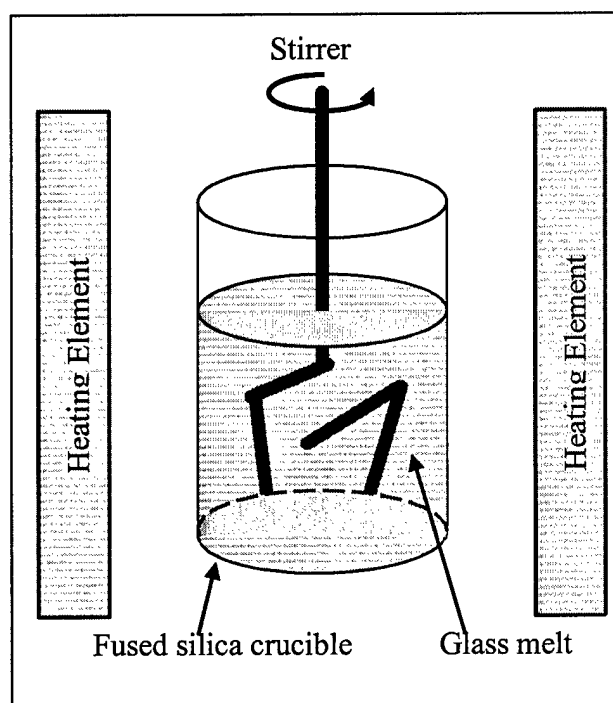


Fig. 5. PTR glass melting and stirring in a silica crucible.

composition (mol. %)  $15\text{Na}_2\text{O}-5\text{ZnO}-4\text{Al}_2\text{O}_3-70\text{SiO}_2-5\text{NaF}-1\text{KBr}-0.01\text{Ag}_2\text{O}-0.01\text{CeO}_2$  was studied. The batch for glass melting was prepared of high purity chemicals and underwent mixing in the shaker-mixer Glen Mills type T2C. As a result, we had the homogeneous batch which was used for glass melting. The glass was melted in an electrical furnace DelTech model DT-31-RS-OS in 400 ml fused silica crucibles at  $1460^\circ\text{C}$  for 7 hours. To provide an optical homogeneity of the glass boule, the multiple stirring of the melt was applied for 2-3 hours at the rate no higher than 20 rpm (Fig. 5). A special shape quartz stirrer, providing good mixing of glass, was customized especially for these melts. After the melting, the glass was cooled down in a crucible on air. The crucible with glass was placed at the massive metal slab that enables to reduce temperature very quickly down to  $T_g$  to avoid a spontaneous crystallization of glass. A spontaneous crystallization occurs quite often while the temperature of cooling glass is passing the region of crystallization from  $700^\circ\text{C}$  to  $550^\circ\text{C}$ . Further the glass boule was undergone annealing with a cooling rate  $0.1^\circ\text{C}/\text{min}$  in the region of structural and stress relaxation in a Lindberg/BlueM box furnace.

To provide the high homogeneity of the glass blanks, we had to reject the traditional casting because it was impossible to prevent the lamination of different parts of the glass melt having different compositions (Fig. 6). This effect is caused by the presence of the components with high volatility in the PTR glass composition. These components are fluorine, bromine, sodium, and silver. The high volatility of the said components results in occurring of the glass layer with the lack of these components on the top of the glass melt. The second process resulting in a creation of striae is the dissolution of the silica crucible and the stirrer in the process of glass fining. In this case of cooling the melt in the crucible, the structure of inhomogeneities in the glass volume is frozen and conserved. We have found that the main areas of inhomogeneity are the outside layer next to the top, bottom and vertical walls of the crucible with the thickness of about 3 mm and an irregular

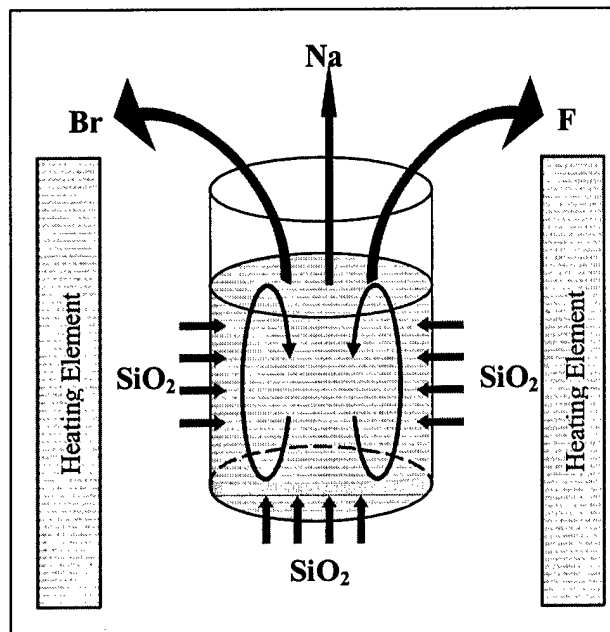


Fig. 6. Sources of optical inhomogeneity in the PTR glass melting process

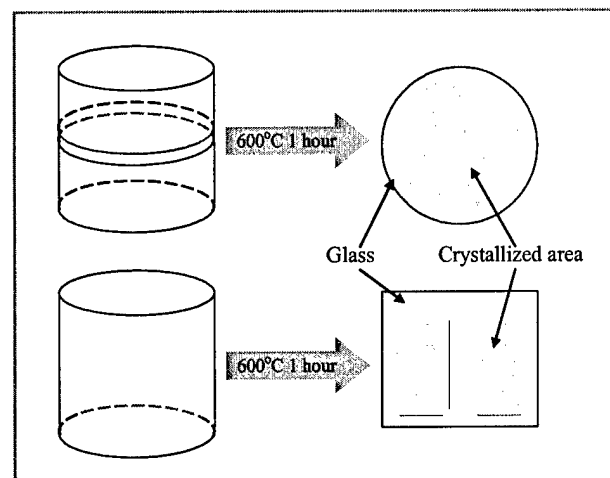


Fig. 7. Method of selection of the PTR glass blanks with homogeneous distribution of refractive index and photosensitivity.

vertical column in the center of the glass boule. Thus, the glass boule was sliced in the horizontal direction and three marked slices from the top, middle, and bottom areas were crystallized at the furnace at 600°C (Fig. 7). The projections of the areas with the low rate of crystallization were selected in the rest of the slices, and the glass plates with the size of 25 mm by 25 mm were cut off.

Annealed glass was cut in slices with thickness 2 mm using the saw Buehler mod. Isomet 1000. Then samples 25 mm by 12 or 25 mm were polished on variable speed grinder/polisher Buehler mod. Ecomet 3. The roughness of a surface of the polished glass specimens were controlled by a profilometer New View 5000. The regular quality of our polishing provided the roughness value ranged from 1 nm to 2 nm. The flatness of the glass blanks was measured by the testing plate. The regular flatness of our samples was about 0.5 wavelengths. The first test for optical homogeneity of the glass blanks was produced by using the Schlieren method. The refractive index mapping and the photosensitivity measurements were provided by the special liquid-cell shearing interferometer.

Finally, the described technology provided a fabrication of PTR glass blanks with size of 25 mm×25 mm with thickness ranged from 0.5 mm to 5 mm. The refractive index fluctuations were below  $10^{-5}$  at the spatial scale of 5 mm and below  $10^{-4}$  at the spatial scale of 25 mm. The photosensitivity fluctuations were below 15 % for the same glass melt and below 25 % for the different melts. This means that the homogeneous photosensitive glass technology enabling the demonstration of the PTR diffractive optical elements was developed and the samples for the project were fabricated.

### **3. New architecture of the PTR glass melting facility**

The main goal of this project was creation of a glass melting facility, which will enable for fabrication of photo-sensitive optical quality PTR glass blanks up to 35×35 mm with refractive index change not exceeding  $10^{-5}$  across such area. Several requirements must be realized for achieving such results. First of all, according to a conventional optical glass technology, the amount of a melt has to be in the range of a few liters. In this case, continuous melting, stirring, and refining secure a fabrication of a homogeneous glass. Smaller volume of the glass melt does not allow high quality of optical glass having no striae and bubbles. Moreover, the PTR glass, which contains volatile and polyvalent elements, makes requirements to the glass melting equipment even more complicated. One of the main sources of striae is change of glass composition during melting resulted from exchange of fluorine (a component of glass melt) with oxygen from the air. To avoid creation of such striae, it is necessary to prevent any contacts of the melt with the air. The next difficulty in the PTR glass melting is keeping of stable equilibrium between polyvalent elements. Any changes in environment, heating conditions, and a composition of glass can result in disturbance of the equilibrium, which plays significant role in photo-chemical processes during a hologram recording. Taking in account all those problems and restrictions, we have analyzed many existing glass melting furnaces and glass melting technologies in order to create the facility, which meets our requirements. The following design of the future melting facility was developed.

## Specification for the PTR melting setup

### Goal of the setup

fabrication of PTR glass blanks with diameter 50 mm and thickness up to 1 cm having refractive index fluctuation across the aperture less than  $5 \times 10^{-6}$  (5 ppm), total contamination with iron and other transition metals below 1 ppm, no platinum inclusions.

### PTR glass properties

Composition (mol. %):  $15\text{Na}_2\text{O}-5\text{ZnO}-4\text{Al}_2\text{O}_3-70\text{SiO}_2-5\text{NaF}-1\text{KBr}-0.01\text{Ag}_2\text{O}-0.01\text{CeO}_2$

Melting temperature  $1460^\circ\text{C}$

Viscosity: 100 P

### General requirements

Glass melting setup (Fig. 8) should include middle frequency induction furnace providing melting of 1 liter of fluorinated glasses at temperature up to  $1500^\circ\text{C}$  with casting through the downpipe at Pt-crucible with separated heating element, Pt-stirrer, and casting device. Furnace should be installed in a locked box. Power supplies and electric circuits must meet USA requirements (voltage, safety). It would be beneficial if the inductor allows scaling up to 2 liter crucible. Experimental melting should be demonstrated at the manufacturer facilities.

### Crucible (Fig. 9)

Desirable ratio of inner diameter to height of crucible  $h = H/ID = 1.5$ . Height of crucible should be 30 % higher the glass surface. For

cylindrical crucible,  $d = \sqrt[3]{\frac{4V}{\pi h}}$  where  $h = H/d$ . For  $V = 1.3 \text{ l}$ ,  $d = 100 \text{ mm}$ . Then  $H_{cr} = 150 \text{ mm}$ .

Downpipe diameter for glass having viscosity about 100 P and mold diameter about 50 mm, should be about 25 mm. (maybe experimental melting at the manufacturer facilities can be conducted.) Downpipe length should be about 100 mm. Modeling or experimental testing would be useful for determination of the crucible geometry.

Thickness is 1 mm. Volume of Pt is  $P_{Pt} = \pi \rho \left[ d_{cr} H_{cr} + d_{dp} l_{dp} + 0.25 (d_{cr} - d_{dp})^2 \right]$  where  $t$  is thickness,  $l_{dp}$  – length of downpipe,  $\rho$  is density. Estimation for such parameters gives 860 g.

### Stirrer (Fig. 9)

Pt-stirrer should be auger, impeller, or rod type (subject for discussion). At the distance of 140 mm lid with diameter of 90 mm should be welded. (Lid design to be discussed.) An electrical mechanism for lifting and lowering of the stirrer should be installed. Estimation of weight of a stirrer with a lid is necessary.

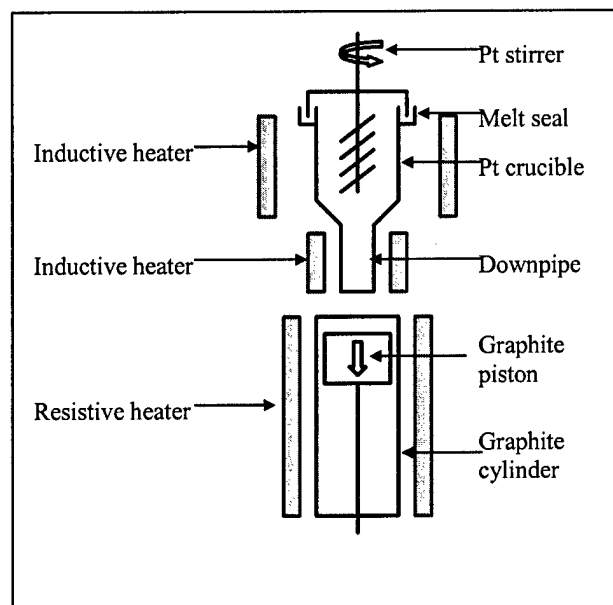


Fig. 8. General design of PTR glass melting-casting-annealing setup

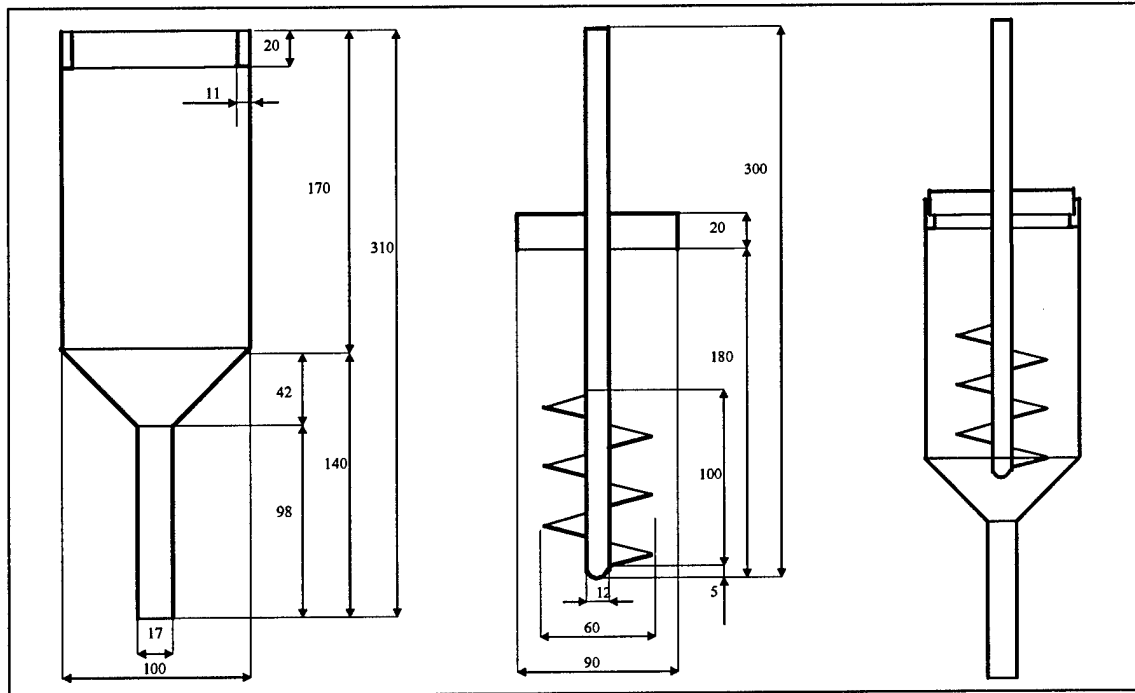


Fig. 9. Design of crucible, stirrer and assembling.

### ***Main inductor***

Inductor should provide enough power to heat 1 liter of glass up to  $1500^{\circ}\text{C}$ , had enough space for the crucible described above. The frequency is to be chosen to provide uniform radial distribution of temperature (probably in the range of 10 kHz, exact number to be determined). Total construction ( It must provide maximum vertical uniformity of temperature field (numbers should be discussed). Temperature in the bottom area should be high enough to prevent crystallization of the melt while downpipe is sealed.

### ***Downpipe inductor***

Downpipe inductor should provide local heating of downpipe for the temperature desirable for homogeneous casting to mold. The frequency range (probably, high frequency) should be discussed. Diameter and length of inductor compare to length and diameter of downpipe are to be determined. A method of temperature control in downpipe should be proposed.

### ***Casting device***

Casting device should include a mold which is a graphite cylinder with diameter of 50 to 60 mm (discuss the exact number), graphite piston sliding inside the cylinder, device for sliding control, and holders for mold and piston. For 50 mm diameter and 1 liter melt, the length of glass casting is about 510 mm. Thus, the mold should have a length about 600 mm. The whole casting device should be placed under the melting device (it should be enough space).

### **Frame**

Frame should hold all devices of the melting setup, have a crucible positioned in such manner that operator can easily operate with it. Total height should not exceed 2,400 mm. The volume with platinum elements should have a locker.

### **4. Creation of the induction melting facility**

Four companies, producing glass and metal melting equipment, such as Deltech, Inc., Jodeit GmbH, Pillar Induction, and Inductotherm, had been involved in this process. Great help we have received from the University of Jena, Germany, which is the birth place of modern optical glass in the world and still have been recognized as a world leader in glass science. Their experience in the glass melting, which they are accumulating for decades, helped us to make our decision in designing of a type of a glass melter. Finally we have chosen an inductive furnace with a platinum crucible incorporated in a furnace. The crucible has a down pipe allowing for casting as-melted glass directly from the crucible into annealing device. Expenditures for the project are summarized in Table 1. It should be noted that final cost the melting setup is higher than it was planned in the beginning. Partially this setup was funded by the DARPA contract "Improved technology for larger aperture Bragg deflectors in PTR glass" which is a part of THOR program.

Table 1. Expenditures for the project account.

Listing of PR/PO by Account Number		650-16-062						1 - Exp
PR#	PO#	Total	Vendor	DATE	Basic Description	Date paid	Date Enbrd	
<i>12-May-04</i>								
<b>099026</b>								
	D05461	\$13,616.00	Pillar Industries	12/23/2002	Two zone melting system as per attached	10/03		2
	D05461	\$91.28	Pillar Industries	11/17/2003	Two zone melting system as per attached	11/03		1
	D05461	\$1,089.01	Pillar Industries	12/15/2003	Two zone melting system as per attached	12/03		1
	D05461	\$3,503.89	Pillar Industries	1/13/2004	Two zone melting system as per attached	01/04		1
	D05461	\$40,848.00	Pillar Industries	12/23/2002	Two zone melting system as per attached	03/03		2
	D05461	\$81,696.00	Pillar Industries	12/23/2002	Two zone melting system as per attached	08/03		2
	D05461	\$750.00	Pillar Industries	12/23/2002	Two zone melting system as per attached	09/03		2
		<b>\$141,594.18</b>						
<b>099152</b>								
	D06538	(\$13,716.80)	ALFA AESAR/A Johnson Ma they Co.	4/14/2004	ID Transfer to 65016069 - Custom Fabrication Pt/5% Ir Crucible/Lid/Stirrer as per drawing			2
	D06538	\$29,352.34	ALFA AESAR/A Johnson Ma they Co.	4/23/2003	Custom Fabrication Pt/5% Ir Crucible/Lid/Stirrer as per drawing	10/03		2
		<b>\$15,635.54</b>						
<b>10671</b>								
	9530	\$6,285.00	Thermcraft, Inc.	9/17/2003	SPLIT TUBE FURNANCE	11/03		2
	9530	\$108.00	Thermcraft, Inc.	9/17/2003	SPLIT TUBE FURNANCE	11/03		1
		<b>\$6,393.00</b>						
<b>Grand Total:</b>		<b>\$163,622.72</b>						

### **Glass melter**

After the bidding process, the glass melting furnace had been ordered to Pillar Induction Company in accordance with our specifications. The furnace is designed for induction glass melting. It is a two zone melting system with independent temperature control for each zone. The

glass is melted by heating a Pt crucible using a MK6S power supply, which is designed to minimize frequency drop-off, at low power levels in order to keep platinum degradation to a minimum. The glass is heated to melting temperature and stirred. Once the mixing process is completed, the second power supply, MK20, turns on in order to heat the downpipe, which allows the molten glass to be cast from the crucible.

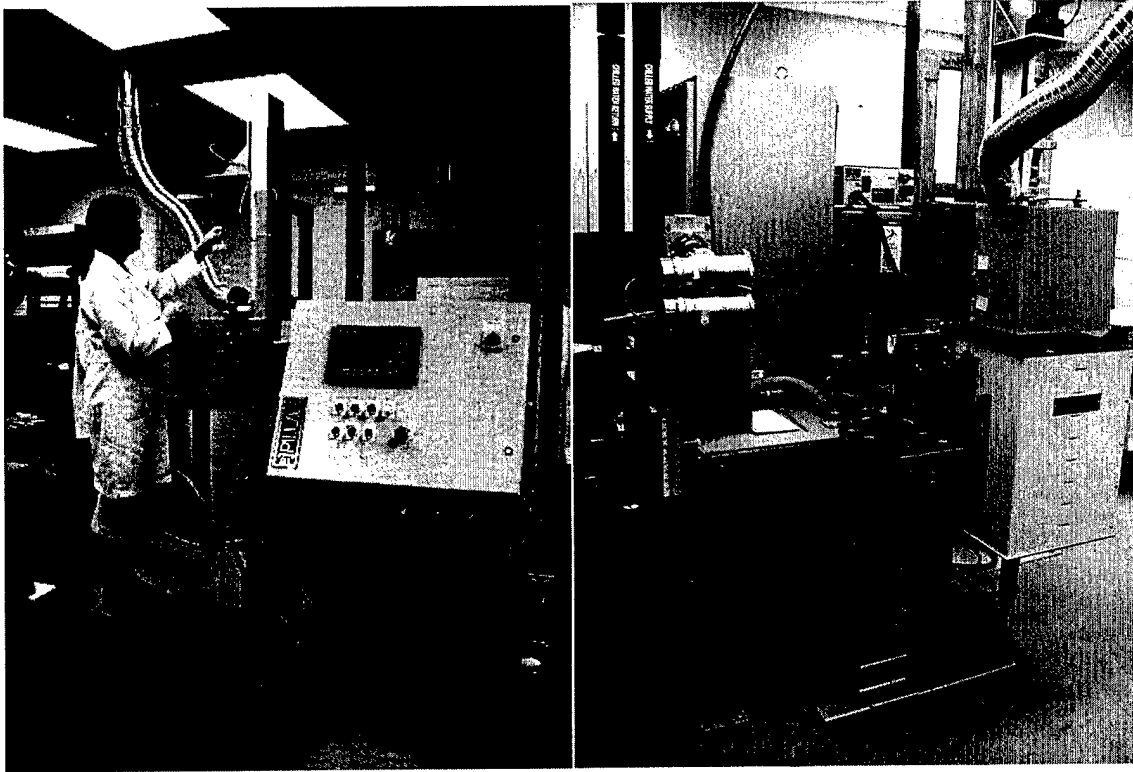


Fig. 10. Glass melting-stirring-casting-annealing setup.

The induction machine is composed of five major components (Fig. 10):

1. Operator control panel, which is located to the side of machine in the electrical enclosure and includes a main disconnect of the electrical control, pushbuttons/switches, and operator interface screen. The Programmable Logical Controller (PLC) allows up to four programs with 10 steps each be stored and used for controlling the melting processes.
2. MK6S power supply, to be used for heating the Pt crucible.
3. MK20 power supply, to be used for heating the downpipe of the crucible.
4. Glass melting furnace, which consists of a refractory shell housing two coils for heating the crucible and the pipe separately.
5. Closed loop pressurized cooling system, which is designed to cool both power units. Tower water cools the furnace and the water cooled leads directly and the power supply components are cooled by the closed loop system water.
6. Two zone temperature control system with a PLC to monitor the exterior temperature of the platinum crucible and the downpipe. A PLC is providing overall control of the induction melting system. Two 4-20 mA analog signals are feeding into the PLC to provide temperature monitoring in two heat zones. The temperature signals are generated

by two bicolor infrared sensors. The PLC adjusts a 4-20 mA signals to each induction power supply. These signals are controlling the amount of energy that is delivered to each zone.

This furnace affords us to melt 2.5 kg of PTR glass in 1.5 l Pt crucible with a bottom pouring of as-melted glass.

### ***Platinum glass melting assembly***

The glass melting assembly consists of a platinum 1.2 liter crucible, an auger type platinum stirrer, and a platinum lid, attached to a stirrer shaft. This assembly had been done at Alfa Aesar according to our specifications (Fig. 9). Based on our requirements of glass melting conditions and a glass composition, an alloy of 95% of Pt and 5% of Ir had been chosen. After consulting with manufacturers of the assembly and the glass melting furnace, the wall thickness and the shape of the crucible and the stirrer were finally designed. As you can see on the drawing, the crucible has an inner pocket, which goes around a top of the crucible. After inserting the stirrer inside the crucible, the lid is inserted in the pocket, which is filled with melted glass. Thus, finally we have completely sealed assembly, which does not allow any access of air in the melt. To avoid too high pressure in the crucible, it has a ventilation 1 mm hole in the lid. The crucible has a downpipe, which allows as-melted glass to be released from the crucible after melt directly to the graphite mold for annealing.

The crucible is incorporated in the furnace (Fig. 11) and has several insulation layers between the crucible and the coil.

### ***Stirring mechanism***

Platinum stirrer is attached to a reversible DC motor, which, in turn, is attached to a slide (Fig. 10). This stirring device is located above the glass melting furnace. The motor can be moved up and down along the slide that allows for inserting the stirrer inside the crucible and removing it out. The speed and direction of stirring of the glass melt can be controlled by motor controller. This device enables for stirring of the glass melt in both, clockwise and counter clockwise, directions with the speed in the range from 10 through 60 rpm.

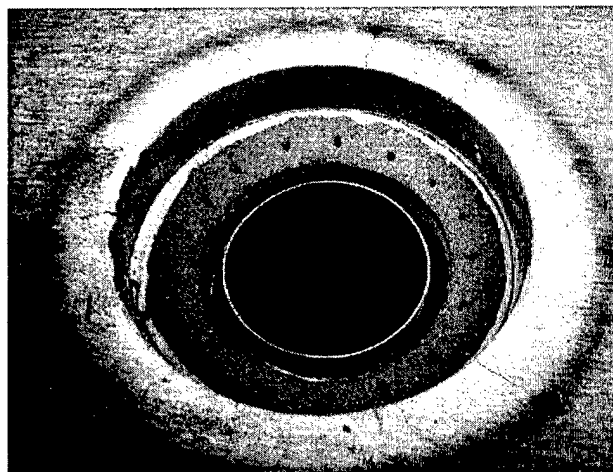


Fig. 11. Top view of the crucible imbedded in the thermo-isolation of the induction furnace.

### ***Casting/annealing device***

A casting device consists of a graphite tube and a piston, which can be moved inside the tube by a DC reversible motor (Fig. 10). A speed and a direction of the piston's movement can be controlled by a motor controller. The graphite tube is placed in the annealing furnace. This split tube furnace, installed in a vertical position, could be open in order to insert or remove the graphite tube. This assembly is placed under the glass melting furnace and centered according to the crucible position. Thus, when as-melted glass comes out of the crucible's downpipe, it goes directly to the graphite cylinder for annealing. In the beginning of the casting the graphite piston

is placed in the upper position inside the cylinder. As soon as glass appears from the downpipe, the piston moves down the cylinder and glass pours in it. The graphite cylinder can be pre-heated to avoid a thermal shock of the cylinder.

## 5. Development of PTR glass melting technology in inductive furnace.

To create the technology of melting the PTR glass in an inductive furnace, we used our experience of melting such glass in resistive furnaces, and also the experience of German scientists of melting fluorine-containing phosphate laser glasses in an inductive furnace. Based on such experience, two step glass melting procedure has been chosen for our experiments. According to this technology, first melt, or so-called cullet melt, conducted in a resistive furnace, in a fused silica crucible, using mix of chemicals. After melting and annealing of this glass, pieces of that melt can be used for the second melt in the platinum crucible in the inductive furnace. This way of re-melting of cullet is widely used in order to get homogeneous glass. As was mentioned above, a very complex composition of glass, containing volatile components and polyvalent elements, results in unstable behavior of the glass in different stages of glass fabrication. So, after creating of the appropriate glass melter, the next step was creating an appropriate glass melting technology. Several melts were conducted in the new furnace following the next steps:

1. Pieces of glass, melted in a resistive furnace, were placed in the platinum crucible and melted at elevated temperature. After all pieces were melted, the stirrer was inserted in the melt, and it was stirred during one hour at the temperature  $1460^{\circ}\text{C}$  to get good homogeneity. As it was described above, the melter was completely sealed and no any contact with air was allowed.
2. Then the downpipe temperature was raised up to  $1460^{\circ}\text{C}$  and glass started pouring down from the crucible. The glass melt, cast in to the graphite tube, then was annealed in the tube, which was placed in the annealing furnace.
3. After annealing, a cylinder of solidified glass could be easily taken out of the graphite cylinder and sliced in desired pieces for a further grinding and polishing.

The next drawbacks have occurred during first three melts:

1. The first melt, which was cast in the cold graphite cylinder, was fully crystallized (left rod in Fig.12)

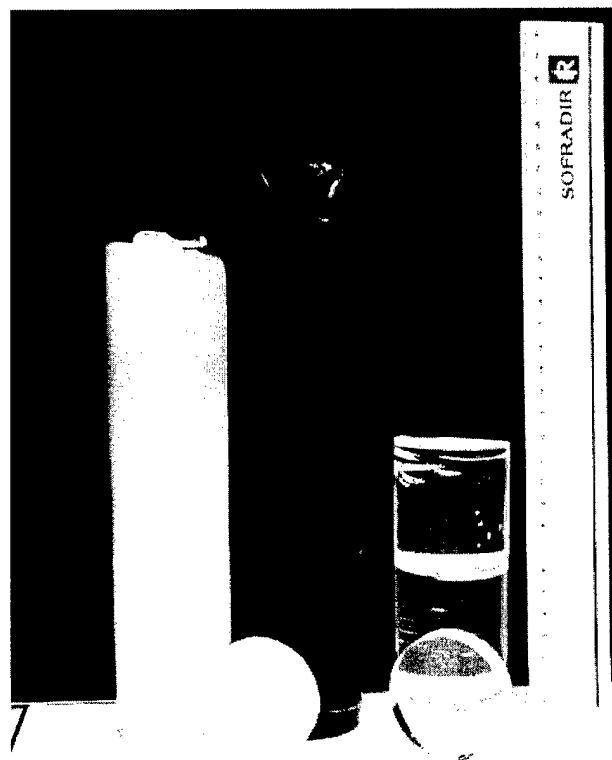


Fig. 12. Castings of PTR glass produced in inductive furnace.

2. The melt, which was done in the completely sealed crucible, had a brown color due to reduced silver (Central rod in Fig. 12).
3. Several melts had a spiral pattern on a cross-section, which can be detected by a shadow method. It was caused by a spiral movement of glass flow coming out of the downpipe.(Fig. 13)

To get over those drawbacks we made the following changes in the glass melting and annealing processes:

1. The graphite cylinder, which serves as a mold for the glass cast, was preheated up to the temperature lower than  $T_g$  of PTR glass. This temperature was determined by several experiments because too high temperature of a mold leads to a spontaneous crystallization, whereas too low temperature results in crack of the solidified glass. To avoid a temperature gradient along the cylinder, which results in glass crack, more reliable thermo insulators were used for the annealing furnace. It allowed us eliminating the thermal gradients and an overheating, which occur during casting glass and further heating glass for annealing.
2. The reduction of silver can be eliminated by changing of a glass composition or by increasing of contact of the air and the glass melt. Second approach is not desirable because it deteriorates the homogeneity of glass. But, on the other hand, the glass composition change can dramatically modify optical properties of the glass. We have solved that problem by the partial increasing of the contact of the glass melt with the air, but we think it is not the last decision and must be studied in the future.
3. The spiral pattern on the cross-section of the glass was eliminated by minimizing a distance between the downpipe and the graphite mold. Also we decreased the temperature of the casting glass in order to increase its viscosity. In this case, the glass flow becomes laminar and the spiral inhomogeneities were diminished. The right rod in Fig. 12 does not show any coloration or scattering.



Fig. 13. Casting of PTR glass from inductive furnace to a steel slab.

Fig. 14 shows polished blanks of PTR glass after casting to the graphite cylinder with diameter of 50 mm. One can see that original glass is homogeneous having no coloration or scattering. The left sample was crystallized at temperature of 600°C to study uniformity of a crystallization rate. One can see that homogeneous white scattering in a crystallized sample proves uniformity of photosensitivity of the PTR glass produced by means of new technology.

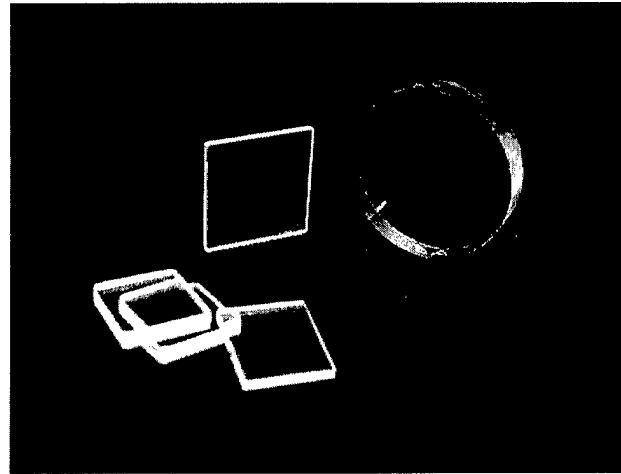


Fig. 15. Polished PTR glass slabs 25×25 mm<sup>2</sup>, 35×35 mm<sup>2</sup> and OD-50 mm.

Fig. 15 shows polished slabs of PTR glass with shapes that are used in the projects for hologram recording rectangular plates 25×25 mm<sup>2</sup> and 35×35 mm<sup>2</sup> and round plates with diameter of 50 mm. Refractive index homogeneity was measured in these samples. It was found that refractive index fluctuation do not exceed 10 ppm at 90% of diameter of the round sample. This means that productivity of PTR technology increased about two times compared to the current technology. Photosensitivity in the last melt has shown 1000 ppm refractive index change that is the highest sensitivity for PTR glass.

Thus, the project successfully completed and new technology enables fabrication of large aperture high-efficiency diffractive optical elements for high power laser beam control.

## 6. DoD projects enabled by new melting facility

- Improved technology for larger aperture Bragg deflectors in PTR glass. DARPA/THOR.
- Incoherent combining of high-power laser beams by volume diffractive gratings in a photosensitive silicate glass. AFRL.
- Volume diffractive optical elements for high-power laser beam control. ARL.
- Photo-thermo-refractive Bragg mirrors for high efficiency diode sources. DARPA/SHEDS.
- High efficient volume holographic optical elements for high power lasers. DARPA/STTR/Light Processing and Technologies, Inc.
- Bragg selectors of transverse modes for high power lasers. MDA/STTR/Light Processing and Technologies, Inc.

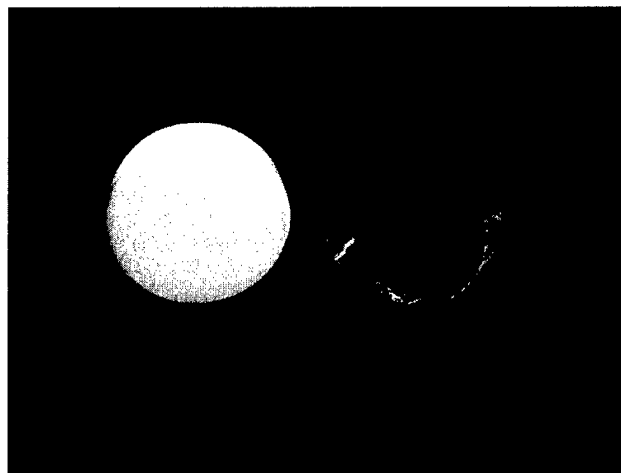


Fig. 14. Homogeneous pieces of PTR glass produced in the inductive furnace. Diameter 500 mm. Left piece is crystallized at 600°C.

One can see from the list of current projects that the new melting facility is immediately used for hologram fabrication which is necessary for a number of projects funded by different services of DoD. It is important to know that this technology is transferring to a company which is spin-off from University of Central Florida. This means that the developed technology very soon will be transformed to commercial technology which would enable supplying not only research projects but large scale fabrication of volume diffractive elements for military application. The use of this technology for commercial market should increase the volume of fabrication and decrease the cost of this new material. A number of preparing proposals for holographic optical elements development will be based in this technology.

## 7. Publications and presentations which use the developed experimental setup

1. I. Ciapurin, V. Smirnov, and L. Glebov. High-density spectral beam combining by thick PTR Bragg gratings. Proceedings of the conference "Solid State and Diode Laser Technology Review" Albuquerque, NM, 2004.
2. L.B. Glebov, L.N. Glebova, V.I. Smirnov, M. Dubinskii, L.D. Merkle, S. Papernov, A.W. Schmid. Laser Damage Resistance of Photo-Thermo-Refractive Glass Bragg Gratings. Proceedings of the conference "Solid State and Diode Laser Technology Review" Albuquerque, NM, 2004.
3. George Venus, Vadim Smirnov, Leonid Glebov, and Manoj Kanskar. Spectral stabilization of laser diodes by external Bragg resonator. Proceedings of the conference "Solid State and Diode Laser Technology Review" Albuquerque, NM, 2004.

## 8. Conclusions

- A unique glass melting facility is created that allows fabricating of homogenous multicomponent silicate glass.
- The uniform PTR glass rods with a diameter of 50 mm and length of 500 mm were fabricated.
- PTR glass blanks of  $35 \times 35 \text{ mm}^2$  was fabricated with uniform photosensitivity and refractive index fluctuations below 10 ppm at 90% of diameter.
- This results pave the way for large aperture precise diffractive optical elements fabrication for optical target characterization and high power laser beam control.

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