

# Development of continuous glass melting for production of Nd-doped phosphate glasses for the NIF and LMJ laser systems

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## ABSTRACT

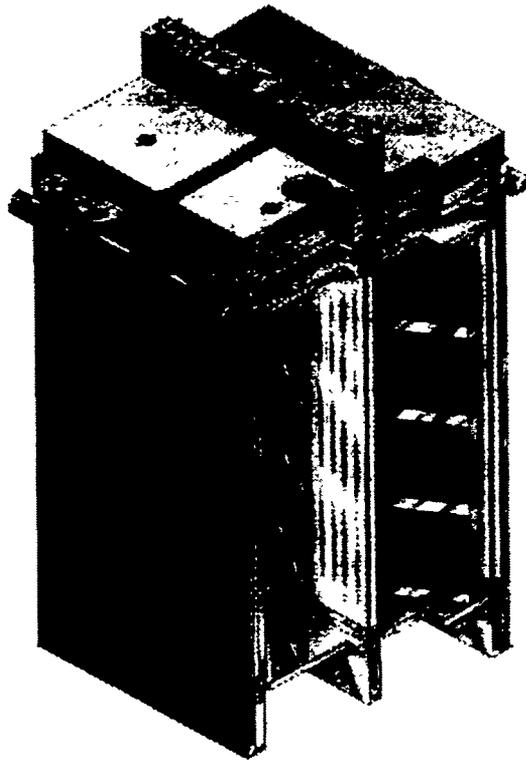
The NIF and LMJ laser systems require about 3380 and 4752 Nd-doped laser glass slabs, respectively. Continuous laser glass melting and forming will be used for the first time to manufacture these slabs. Two vendors have been chosen to produce the glass: Hoya Corporation and Schott Glass Technologies. The laser glass melting systems that each of these two vendors have designed, built and tested are arguably the most advanced in the world. Production of the laser glass will begin on a pilot scale in the fall of 1998.

## 1. INTRODUCTION

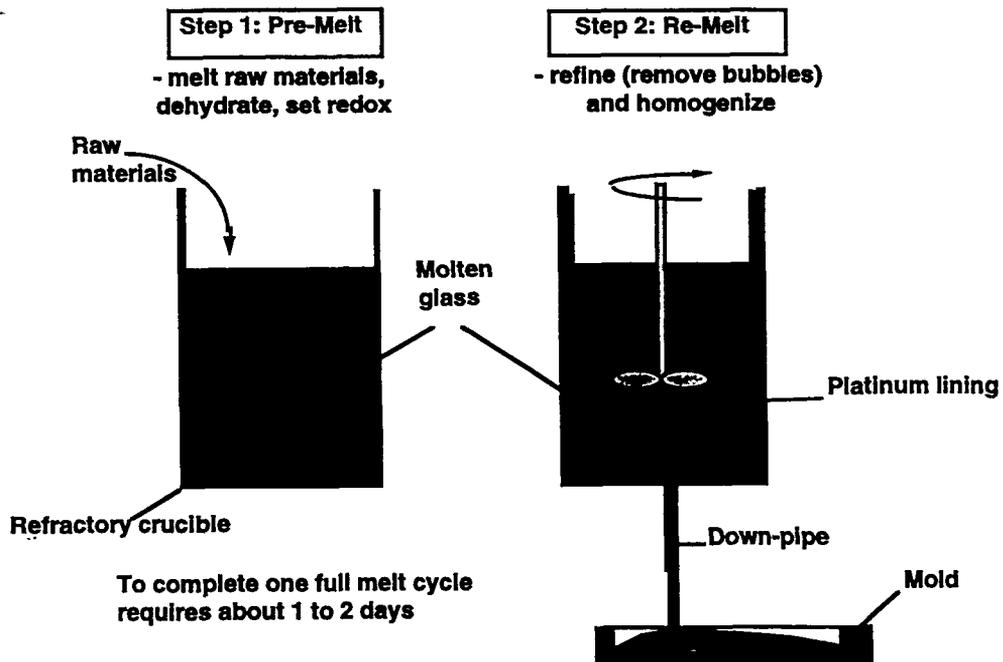
The laser systems that comprise the US Department of Energy (DOE) National Ignition Facility (NIF) and the French Commissariat à l'Energie Atomique (CEA) Laser Megajoule (LMJ), each consist of 192 and 240 laser beamlines, respectively.<sup>1-4</sup> Each beamline contains either 16 (NIF) or 18 (LMJ) large, Nd-doped laser glass slabs and each finished laser glass slab is about  $81 \times 46 \times 4.1$  cm<sup>3</sup>. A total of 3072 and 4380 slabs will be installed on the NIF and LMJ, respectively (Table 1). In addition, each facility plans to purchase approximately 10% more slabs as construction and operation spares. Thus, nearly 8150 laser glass slabs will be needed for the two laser systems: this represents a volume of about 125 m<sup>3</sup> (330 metric tons) of *finished* high optical quality glass. Note that the quantity of raw glass that must be melted is significantly larger than 330 metric tons to account for various processing losses.

Both the NIF and LMJ use a compact laser amplifier design called the "multi-segment amplifier" (MSA).<sup>5,6,7</sup> These amplifiers consist of stacked  $4 \times 1$  arrays of laser glass slabs inside a flashlamp pumped cavity (Fig 1). By using square apertures (i.e. square beams) it is possible to tightly pack the individual laser glass amplifiers into a compact matrix and greatly reduce the size and cost of the system. This design requires that the laser glass be manufactured in rectangular slabs. Note that although the laser aperture is square, the laser slabs are rectangular because they are mounted at Brewster's angle to the propagation direction of the beam. Mounting the glass at Brewster's angle minimizes the Fresnel reflection losses at the surfaces of the slabs. In addition, mounting at an angle increases the coupling efficiency of the flashlamp light with the slabs. Erlandson et al.<sup>5,6,7</sup> have recently described in detail the design and operating characteristics of flashlamp pumped multi-segment amplifiers. The measured small signal gain coefficient is typically about 0.05/cm and the stored energy density about 0.25 J/cm<sup>3</sup> for phosphate laser glass doped at about  $4 \times 10^{20}$ /cm<sup>3</sup> and pumped at a lamp explosion fraction of 0.20.

A prototype laser closely resembling one of the NIF beamlines has recently been built and tested.<sup>8</sup> This laser, called "Beamlet", uses 11 amplifiers in the main cavity and 5 in the booster section, the same configuration as the NIF. A series of large phosphate glass slabs ( $767 \times 428 \times 44$  mm<sup>3</sup>) having a doping of  $3.5 \times 10^{20}$ /cm<sup>3</sup> were produced for this laser.<sup>9</sup> Although slightly smaller than required for LMJ and NIF, these prototype glass slabs were made to nearly identical specifications as required for the NIF and LMJ. Therefore, to a great extent, the quality and size of the laser glass pieces needed for NIF and LMJ have been demonstrated. What remains is to develop the manufacturing capability for producing a large number of these high quality glass slabs at a high rate and at a significantly lower cost. In this paper we briefly describe the advanced melting methods that are being developed to produce the glass for the NIF and LMJ. Although details of the production processes are proprietary, we will highlight the changes involved in changing from the one-at-a-time discontinuous production process used in the past to the continuous melting process of the future. Our cost goal is to manufacture the laser glass for about \$1000/liter (\$350/kg); this is roughly a factor of three lower than the cost with the current one-at-a-time production methods. Given the size of the laser glass order, this level of cost reduction represents a total cost savings of about \$200 to \$300 million over the price that could be achieved with current manufacturing methods.



**Figure 1.** Assembly drawing of two  $4 \times 2$  amplifier units to be used on NIF. The laser slabs are mounted in the amplifiers in a frame that contains four (4) vertically stacked laser slabs.



**Figure 2.** Schematic representation of the current discontinuous, 2-step process used to melt and form laser glass castings.

Table 1: Quantity of laser glass required for the LMJ and NIF projects.

<u>Variable</u>	<u>NIF</u>	<u>LMJ</u>
Number of beamlines	192	240
Number of slabs / beamline	16	18
Spares	10%	10%
Total number of slabs (including spares)	3380	4752
Finished slab dimensions (cm <sup>3</sup> )	81 × 46 × 4.1	81 × 46 × 4.1
Volume per slab (liter)	15.3	15.3
Total volume (m <sup>3</sup> )	52	73
Mass (metric tons)	136	190

## 2. LASER GLASS SPECIFICATIONS AND PROPERTIES

### 2.1 Optical quality

Some of the most critical specifications of the laser glass relate to its optical quality and are strongly dependent on the processing conditions. In particular, there are three main characteristics of the glass that impact optical quality:

- optical homogeneity
- inclusions
- bubbles

Optical homogeneity refers to the refractive index variation in the optical material and for laser glass this is typically less than 2 ppm (i.e.  $\Delta n < \pm 2 \times 10^{-6}$ ). The homogeneity is generally specified in terms of a maximum amount of allowed aberration due to sphere, coma, astigmatism and a smaller amount of higher-order terms (see Table 2). For NIF and LMJ we intend to keep this same specification. However the final, finished (that is, polished) laser glass will be specified using a more sophisticated procedure designed to monitor aberrations at specific spatial frequencies that are known to seed non-linear-growth of intensity noise in the laser beam.

The homogeneity of the laser glass is critical in order to maintain wavefront uniformity of the laser beam. Recall that there are a total of 16 laser slabs in the laser beamline and during the four passes through the amplifier chain the beam passes through the equivalent of 64 laser slabs. Therefore, even small optical inhomogeneities can lead to significant wavefront aberration in the output of the beam, potentially causing significant degradation in both frequency conversion and focusability of the beam.

Inclusions from ceramic refractory materials, unmelted raw materials, Pt metal, crystallites or impurities can cause optical damage in the glass when exposed to high laser fluences. The most common inclusion source is metallic Pt inclusions from the Pt liners used in the melting system. Improved processing conditions have led to a dramatic reduction in Pt inclusions in recent years such that the average inclusion density is less than 0.1 per liter of glass or less than an average of 1 to 2 per glass slab.<sup>10,11</sup> Inclusions in the laser glass typically damage at about 2–5 J/cm<sup>2</sup> at the NIF and LMJ pulse lengths.<sup>12</sup> Although very small to begin with, inclusion damage can grow with successive laser shots to several millimeters or even centimeters in size making the laser glass unusable. Also large damage spots in the laser glass can seed damage in other optics in the laser chain. In general, if the inclusions are small they can be tolerated as long as the optical damage they produce does not exceed about 250  $\mu\text{m}$  in size. This is the basis for the specification given in Table 2. Currently we scan each piece of laser glass with a high fluence laser beam and measure the size of any damage site after a specified number of shots at fluences between 7 to 14 J/cm<sup>2</sup> (8 ns).<sup>13</sup> If the Pt-damage size remains below the specified size limit given in Table 2, then it is acceptable.

The laser glass bubble specification is based on two requirements: first, the need to minimize the amount of light loss due to the obscurations caused by the bubbles and second, the need to keep the size below a certain value that may induce

non-linear growth of intensity noise. The obscuration loss is not to exceed 0.01% of the beam area per slab and therefore sets the total number of bubbles of a given size allowed in any given slab. The maximum size bubble allowed is currently 100  $\mu\text{m}$ . The diffracted light from bubbles that exceed 100  $\mu\text{m}$  can, at high intensities, imprint a holographic diffraction pattern in the next optic that, in turn, can bring that portion of the beam to focus at another downstream optic and potentially damage it. Because of the regular spacing of many of the optics in the laser chain this non-linear imaging effect could lead to propagation of laser damage throughout the beamline. In general, bubbles have not been a significant problem for laser glass. For example, only one of the Beamlet slabs had bubbles and these were so few in number and so small as to be insignificant.<sup>9</sup> Similar results were observed for the Nova and Phebus laser glass disks.

Table 2: Several key technical specifications for the NIF *pre-finished* laser glass slabs. These pre-finished slabs will be clad, ground and polished into the finished slabs by the finishing vendor.

<b>Parameter</b>	<b>NIF specifications</b>
1. Nd doping	$4.2 \times 10^{20} \pm 0.1 \text{ Nd}^{3+} / \text{cm}^3$
2. Homogeneity (expressed as wavefront error at @ 632 nm, normal incidence)	
Sphere	$< 0.425 \lambda$
Astigmatism	$< 0.220 \lambda$
Higher order aberrations	$< 0.142 \lambda$
3. Fluorescence Lifetime (measured on $5 \times 5 \times 0.5 \text{ cm}^3$ sample)	$> 320 \mu\text{sec}$
4. Absorption coefficients:	
at 1053 nm	$\leq 0.0019 \text{ cm}^{-1}$
at 400 nm (due to $\text{Pt}^{n+}$ )	$\leq 0.25 \text{ cm}^{-1}$
at 3333 nm (due to $-\text{OH}$ )	$\leq 2 \text{ cm}^{-1}$
5. Bubbles:	
max. number (per 100 $\text{cm}^2$ area)	total cross section $< 0.15 \text{ mm}^2$
maximum diameter	$\leq 100 \mu\text{m}$
6. Birefringence	$\leq 5 \text{ nm/cm}$
7. Pt inclusions:	
max. number for any one slab	$\leq 5$ in Clear Aperture
average for all slabs	$\leq 2$ per slab
max. size after laser irradiation	$\leq 100 \mu\text{m}$ .

## 2.1 Properties of the NIF and LMJ laser glasses

Laser glasses are specially formulated to give the desired laser, optical, thermal-mechanical and physical-chemical properties needed for a specific laser application. Some of these properties are strongly affected by the processing conditions (as discussed in the preceding sections), however, most are controlled by the base glass composition.

We have chosen two glasses for use on the NIF and LMJ that meet the gain, energy storage, extraction efficiency, and damage resistance requirements: LHG-8 (Hoya Corporation) and LG-770 (Schott Glass Technologies Inc.). LHG-8 is the same glass used on the Nova and Phebus lasers, however, LG-770 is a new formulation developed to replace LG-750 that was used on Nova, Phebus and Beamlet.

The key properties of these laser glasses are summarized in Table 3. The importance of these properties for ICF high-peak-power applications has been recently reviewed.<sup>14</sup> Consequently, we will not discuss laser glass properties further here.

Table 3 : Properties of LHG-8 (Hoya) and LG-770 (Schott), the two laser glasses selected for use on LMJ and NIF.

<b>Glass Properties</b>	<b>LHG-8</b>	<b>LG-770</b>
<b>Optical</b>		
Refractive index		
$n_d$ (589.3 nm)	1.52962	1.50674
$n_1$ (1053 nm)	1.52005	1.49908
Non-linear refractive index		
$n_2$ ( $10^{-13}$ esu)	1.12	1.01
$\gamma$ ( $10^{-20}$ m <sup>2</sup> /W)	3.08	2.78
Abbe number	66.5	68.4
<b>Laser</b>		
Emission cross-section ( $10^{-20}$ cm <sup>2</sup> )	3.6	3.9
Radiative lifetime at zero-Nd concentration ( $\mu$ s)	365	372
Judd-Ofelt radiative lifetime ( $\mu$ s)	351	349
Emission band width (nm)	26.5	25.4
<b>Thermal</b>		
Thermal conductivity, 90°C (W/mK)	0.58	0.57
Thermal diffusivity ( $10^{-7}$ m <sup>2</sup> /s)	2.7	2.9
Specific heat, $C_p$ (J/gK)	0.75	0.77
Coefficient of thermal expansion, 20–300°C ( $10^{-7}$ /K)	127	134
Glass transition temperature, $T_g$ (°C)	485	460
<b>Mechanical</b>		
Density (g/cm <sup>3</sup> )	2.83	2.59
Poisson's ratio	0.26	0.25
Fracture toughness (MPa • m <sup>1/2</sup> )	0.51	0.48
Hardness (GPa)	3.43	3.58
Young's modulus (GPa)	50.1	47.3

### 3. GLASS MELTING DEVELOPMENT

The glasses made for the present large ICF laser systems (e.g. Nova [LLNL], Phebus [CEA], Beamlet [LLNL] Gekko [Osaka] and Omega [Univ. of Rochester]) were manufactured using a one-at-a-time, discontinuous melting process. In this section we briefly describe this older manufacturing method and the new advanced processes that have been developed for NIF and LMJ.

#### 3.1 The "old" technology: one-at-a-time, discontinuous laser glass melting and forming

The first step of the discontinuous process is a pre-melting step designed melt and mix (on a large scale length) the raw starting materials (Fig. 2). A bubbling gas is often added to remove unwanted volatile products, particularly water and, if necessary adjust the melt redox state. The pre-melt is carried out in a relatively inert refractory crucible. The walls of the refractory vessel corrode over time eventually requiring the vessel to be replaced. The glass from the pre-melter generally contains bubbles, striae, and possibly some small particles of unmelted starting material.

The product glass from the pre-melt stage is next processed in a physically separate unit called the "re-melter". The re-melter consists of a platinum lined vessel that also has provisions for stirring and gas bubbling. The main purpose of the re-melter is to dissolve any platinum inclusions, remove any bubbles and finally, homogenize the glass to provide the striae-free, high-optical-quality glass necessary for laser applications. This process involves several stages.<sup>15</sup> During the first stage

of the re-melt cycle the redox state of the glass is adjusted to enhance platinum particle dissolution. This followed by a refining or "fining" process conducted at high temperatures where the viscosity of the glass is low, allowing bubbles to rise to the surface. The third stage is a stirring process which is generally conducted at temperatures lower than either the melting or the refining stages. The continuous stirring thoroughly distributes all components within the glass melt, eliminating striae and thus ensuring uniformity of the refractive index over the entire casting. Finally, the melt is cooled to a temperature such that the viscosity of the glass is proper for casting into a mold of the appropriate size and shape. After casting, the glass undergoes a coarse annealing step, is inspected for inclusions and striae, and then is fine annealed to remove residual thermal stresses due to the forming process.

### **3.2 Advanced technology: continuous laser glass melting and forming**

Advanced laser glass melting processes have been developed separately by Schott Glass Technologies (Duryea, PA) and Hoya Corporation (Fremont, CA) under work funded jointly by the Lawrence Livermore National Laboratory and the Centre d' Etudes de Limeil-Valenton. The two glass companies have chosen different development approaches. Schott has chosen to design and develop a full-scale melting system that will then become the production melter. This approach allowed for one development run to verify the equipment design and the melting and forming process; this was completed in November and December 1997. In contrast Hoya chose to carry out development using a sub-scale continuous melter. Because of the smaller size and lower operating costs of this equipment Hoya was able to carry out several melting and forming campaigns which were completed in March 1998. We are pleased to state that both vendors have successfully completed their development efforts and are preparing for a first production run that we term the "pilot". This will be followed by several years of production, approximately 2-3 years for the NIF and about 3-4 years for LMJ.

Many of the details of the manufacturing process are highly proprietary to each company. Therefore, we give only a generic description of the melting, forming and coarse annealing process. Nevertheless, this description should give an idea of the progress in laser glass manufacturing technology that has occurred as a result of the NIF and LMJ projects. The laser glass melting systems developed by Schott and Hoya are arguably the most advanced optical glass melting systems in the world.

A continuous optical glass melting system is generally divided into several interconnected zones (Fig. 3). Each zone consists of one or more vessels designed to carry out a specific aspect of the process. In the case of the laser glass continuous melters there are six main processing zones<sup>16</sup> (Fig 3): (1) raw material batching, (2) melting, (3) conditioning, (4) refining, (5) homogenization and, (6) continuous strip forming. These regions are interconnected allowing for the continuous flow of glass from one zone to the next.

It is desirable that the raw materials be batched together and then thoroughly mixed in a dry atmosphere. The batch is then delivered continuously to the melter with precautions to avoid water uptake by hygroscopic raw materials. The batch powder that enters the melter dissolves in the molten glass and undergoes large-scale mixing. Off-gas handling equipment collects any gas emissions from the melter (or other vessels) and treats the effluent to meet environmental regulations.

The glass continuously flows from the melter into the conditioning unit where the redox state of the melt is adjusted to enhance Pt-inclusion dissolution. If required, steps may also be taken to remove any excess "water" (i.e. -OH) in the glass. The glass from the conditioning unit then flows to the refiner section where the temperature is generally elevated to reduce the glass viscosity and thereby increase the bubble rise velocity to promote bubble removal.

Finally, the glass from the refiner enters the homogenization section where Pt stirrers thoroughly mix the glass to achieve the part-per-million index homogeneity required for ICF laser applications. Just as in the discontinuous process, the temperature of the homogenizing section is reduced to adjust the glass viscosity to give the desired flow characteristics needed to form a wide, thick, homogeneous strip of glass. The width and thickness of the glass strip produced during the forming operation is greater than any optical glass ever produced prior to NIF/LMJ-driven glass development.

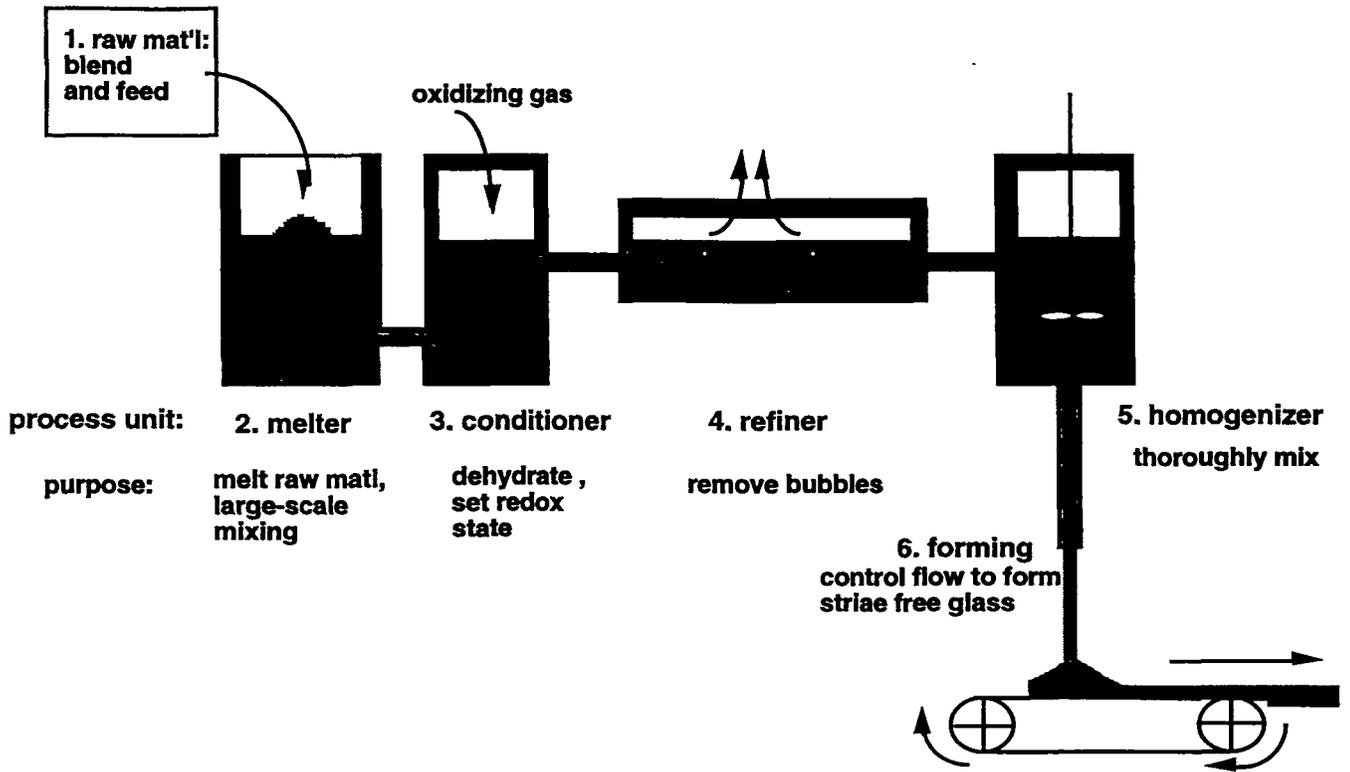


Figure 3. Schematic representation of the continuous laser glass melting systems to be used for manufacture of NIF and LMJ laser glass. Shown are the various process zones that are discussed further in the text.

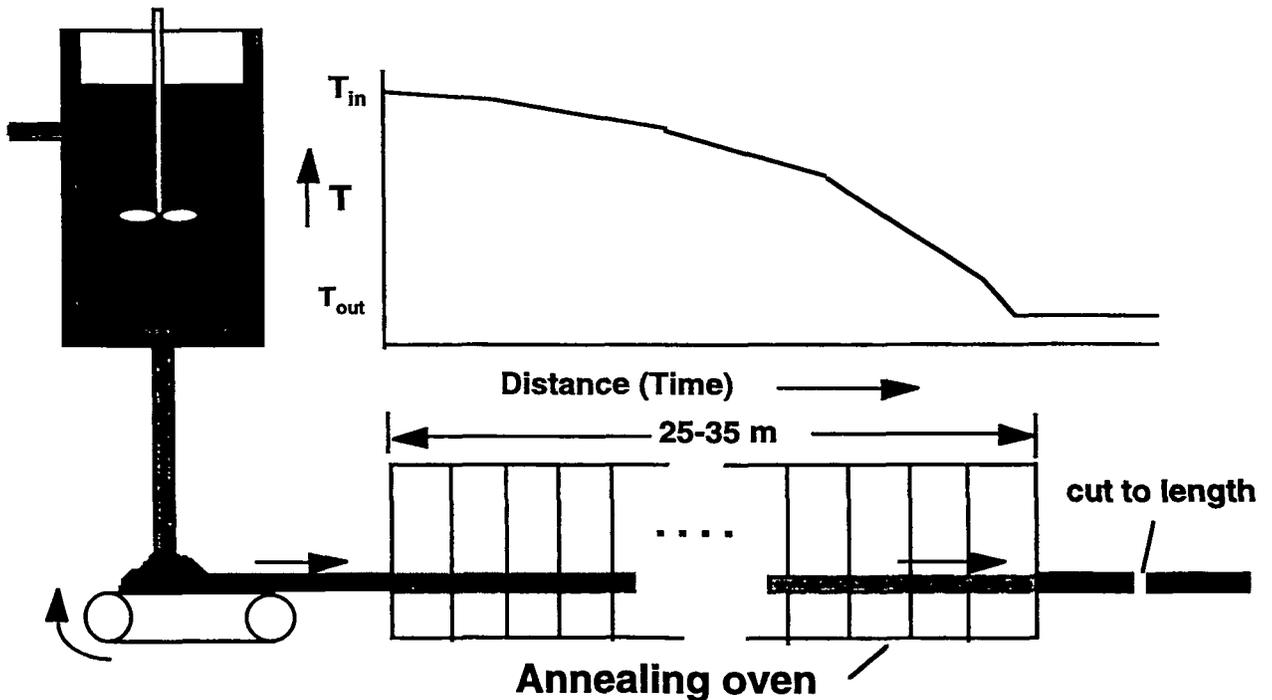


Figure 4. Schematic representation of the system used to coarse anneal the as-cast, continuous laser glass strip.

The glass manufacturers have highly proprietary technology that they employ to "form" (i.e. cast) the glass into a homogeneous continuous strip free of sharp index variations (striae). Once successfully formed, the cast strip moves by conveyor belt through a long (25-35 m) coarse annealing oven (Fig. 4) where the temperature is ramped down at a rate to avoid generating unacceptable thermal stresses in the glass. Finally, at the end of the Lehr, the cast strip is cut into pieces that are then individually post-processed to give the desired laser slab blank.

Both manufacturers will use advanced processing conditions designed to minimize the formation of Pt inclusions in the laser glass. Prior to 1986, Pt inclusion damage represented the major source of damage in laser glass used for high peak power applications. However new processing methods effectively reduce the Pt inclusion concentration by more than 1000-fold to fewer than an average of 1 to 2 per laser glass slab (i.e. less than 0.1 per liter).<sup>9-12</sup>

### 3.3 Post-processing

Once the laser glass has been melted and formed into plates, there remains a number of other process steps before the glass can be shipped to the final finishing vendor. Specifically the laser glass needs to undergo pre-fabrication to a size suitable for inspection for striae and Pt inclusions. Following this the glass is slowly annealed to remove any residual strain; this process alone can take many days. Finally the glass is fabricated to the final dimensions required by the glass finishing, inspected for homogeneity and prepared for shipping.

## 4. MANUFACTURING SCHEDULE

The NIF and LMJ laser glass manufacturing is divided into two main phases: pilot and production. Pilot refers to the first production run; the results from the pilot run will be used to establish yield and costs. In addition, the glass from the pilot run will be used by the finishing vendors to demonstrate the advanced laser glass finishing and polishing methods to be used in final production.

The production phase immediately follows pilot. The first stage of laser glass production will be primarily for the NIF facility because NIF construction will occur earlier than the LMJ. The NIF production will take place over approximately three years at a rate of about 1200 slabs per year. This production will be followed by the LMJ production that will last 3 to 4 years. On the third year of production the NIF and LMJ may overlap somewhat causing a short term increase in the annual production rate.

CEA is building an intermediate facility called the "Integrated Laser Line" (LIL). The LIL will consist of 8 beams identical to those on the LMJ (which contains 240). The first goal of this laser is to validate the design of the LMJ including the performance of the various laser components and optics. Some of the laser slabs produced during pilot operations will be used on the LIL.

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