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Reluctant Glass Formers and Their Applications In Optical Lens Design

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Over ten years ago it was shown that glasses with high index of refraction and some with low dispersion could be produced from a number of pure refractory oxides including the lanthanides by containerless processing. By containerless processing it is possible to minimize surface heterogeneous nucleation and produce larger glass samples of the materials than by other methods. The use of proposed high temperature containerless processing facilities in space will permit the fabrication of benchmark samples of new unique glass compositions for optical property determination as well as for glass formation.

It has been shown that glasses with high refractive indices and large Abbe numbers can be formulated using this technology. These glasses lie in the classical "forbidden" region of the glass map. Preliminary study of the impact of having such unusual glasses available for the lens designer has been made. Results indicate that significant improvements can be realized over the use of only conventional glasses. A cursory survey of a number of nationally recognized lens designers indicated a general agreement that such glasses would be highly desired and could be expected to lead to completely new designs as well as simplifying existing designs.

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The art of optical lens system design is very old. Over a hundred years ago flint glasses (lead-alkali-silicates) were discovered to correct the color aberration of the crown glasses (soda-lime-silicates) making the first achromatic color corrected multi-element lens systems possible. Over the following years demands for better quality lenses led to a multitude of glass compositions with unique combinations of index of refraction and dispersion (inverse of the Abbe number) that have made possible the increasing complicated optical systems.

The performance of optical lenses is highly dependent upon the specific selection of glasses incorporated into the design and the properties of these glasses. Over the years, lens designers have observed that the image quality of most lens configurations significantly benefit from the use of glasses with high refractive index. The difficulty experienced by designers is that the dispersion (variation of refractive index with wavelength) of such glasses rapidly increases as the refractive index increases. Although color correction of lenses is accomplished by using combination glasses having both low and high dispersive characteristics, the difference in refractive index at the reference wavelength often cannot be very great without adversely impacting the potential state of correction of the optical aberrations. In order to exploit the anticipated advantages of using high refractive index glasses, it appears to be generally necessary for high refractive index, low dispersion glasses to be available. The technically refraction plotted vs. Abbe number. New compositions with properties up and to the left with high index of refraction and low dispersion are highly desirable. Moreover, for good correction of secondary chromatic aberration, it is often necessary to have glasses available with anomalous dispersion (non-normal partial dispersion).

Over a decade ago Ralph Happe of Rockwell International advanced the science of optical glasses by demonstrating that by containerless processing, small samples of refractory reluctant glass formers could be produced from a large number of nontraditional glass formers. These glasses with high index of refraction and low dispersion could be produced from a number of pure refractory oxides (including the lanthandes) and from eutectics formed from two or more of the oxides. This was done by laser spin melting appropriate compositions of the oxides which solidified in the short free fall time as amorphous small spheres of glass. The small size of the samples limits the determination of the index of refraction of the material to sufficient accuracy to determine reliable dispersion values but a few compositions, however, were found to have both high index of refraction and low dispersion residing the currently forbidden but desirable region of the optical property diagram. Based on the expected values for index of refraction and dispersion it was proposed that new glasses from regions 1,2 and 3 could be mixed to produce virtually any intermediate property between the three new property regions. Containerless processing makes it possible to minimize surface heterogeneous nucleation and produce larger glass samples of the materials than by other methods. The use of proposed high temperature containerless processing facilities in space will permit the fabrication of benchmark samples of new unique glass compositions for optical property determination as well as for glass formation studies.

First results from a preliminary study of the impact of having such glasses available has been made through the study of several simple and common lens configurations. The result of the computer modeling of these lenses showed impressive enhancement of the optical performance. For example, a Tessar lens was first optimized using four conventional glasses and then redesigned by substituting two "new" glasses in the design. Transverse ray aberrations on-axis and at three off-axis field angles for both the conventional design and the "new" design. The improvement in image quality is evident. Further study of the use of "new" glasses in lens design is being directed towards the determination of specifications of specific optical properties for these glasses that can serve as engineering goals for the materials scientist. A cursory survey of a number of nationally recognized lens designers indicated a general agreement that such glasses would be highly desirable and could be expected to lead to completely new designs as well as simplify existing designs.

Production of these new reluctant refractory glasses could be made possible utilizing containerless processing to prevent the contamination of th high temperature melt but more importantly by the suppression of heterogeneous surface nucleation of crystallization during cooling. If heterogeneous nucleation could be minimized, homogeneous nucleation would not occur till much lower temperatures where the systems would be much less susceptible to crystallization due to the increased viscosity of the melt. Critical cooling rates to form glass could be much smaller than with samples cooled in a mold in contact with a container wall. Glasses that are virtually impossible to form in the confined conditions (in lg of Earth) could be possible to form under containerless conditions (low g of space).

A high temperature containerless processing furnace is necessary in order to investigate these new types of optical glasses. One of the authors (EE) has investigated terrestrial containerless processing methods for the past 13 years. Each method has limitations that restrict the size or type of samples that can be processed. None of the methods has proven useful for containerless processing samples large enough to make the necessary optical property measurements to sufficient accuracy.

Due to the upper temperature limits of existing low gravity containerless processing facilities (1475°C in Single Axis Acoustic Levitator SAAL). The Gallia Calcia system was selected as a low melting (1323°C) model of these refractory reluctant glass formers. It has optical properties within the known region of the diagram but is a "low temperature model" for the new glass systems. It is a reluctant glass former that is without the traditional glass formers. Gallia-Calcia may have unique partial dispersion characteristics. From the limited optical property measurements of this material there is an indication that there may be analmous dispersion in this new glass system which implies more fundamental differences in the partial dispersion of this glass compared with the closest commercial glass. Virtually all glasses from the traditional glass formers are concave up. With Gallia-Calcia the curve is concave down. The new families of noncross bred glasses offer possibilities for unique partial dispersion characteristics which could correct the secondary spectrum.

Of the several hundred compositions of potential refractory reluctant glass formers that have been investigated to date very few compositions can be processed in the existing SAAL. Using one potential system as an example, the niobium calcium titanate ternary has a reasonably large glass formation region through the diagram as determined by laser spin melted samples. Only two small regions are molten below 1500°C.

The high temperature requirement for containerless processing is further increased due to the fact that the melts of these materials must be superheated in order to achieve optimal undercooling conditions. It is expected that the melt should be heated at least 200°C above the thermodynamic melting point to minimize nucleation upon cooling. Obviously the superheat required in order to optimize undercooling would have to be determined for each system.

High temperature containerless processing of non-electrical-conducting melts is required for further development of unique optical glasses. Samples on the order of 5 mm to 1 cm in size are required for optical property measurements. Cooling rates need to be very fast in order to avoid nucleation and crystallization. Few (if any) meaningful experiments can be done with a furnace capability below 1500°C. Temperature capabilities to 2000°C is a minimum requirement for such a furnace with 2200°C being a goal. If the production of these glasses can be proven possible, a revolution in optical glasses could result in completely new optical lens systems.

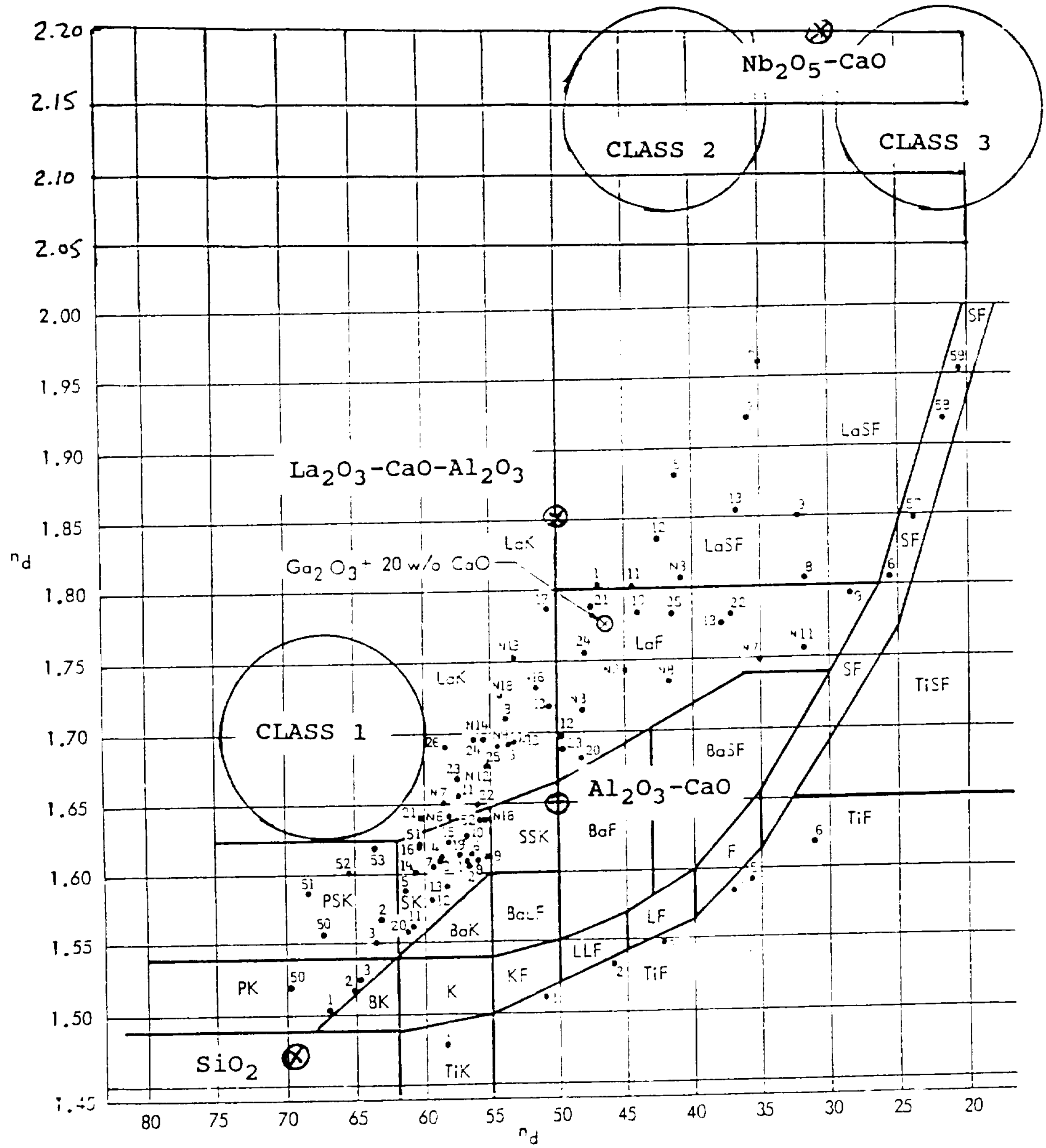
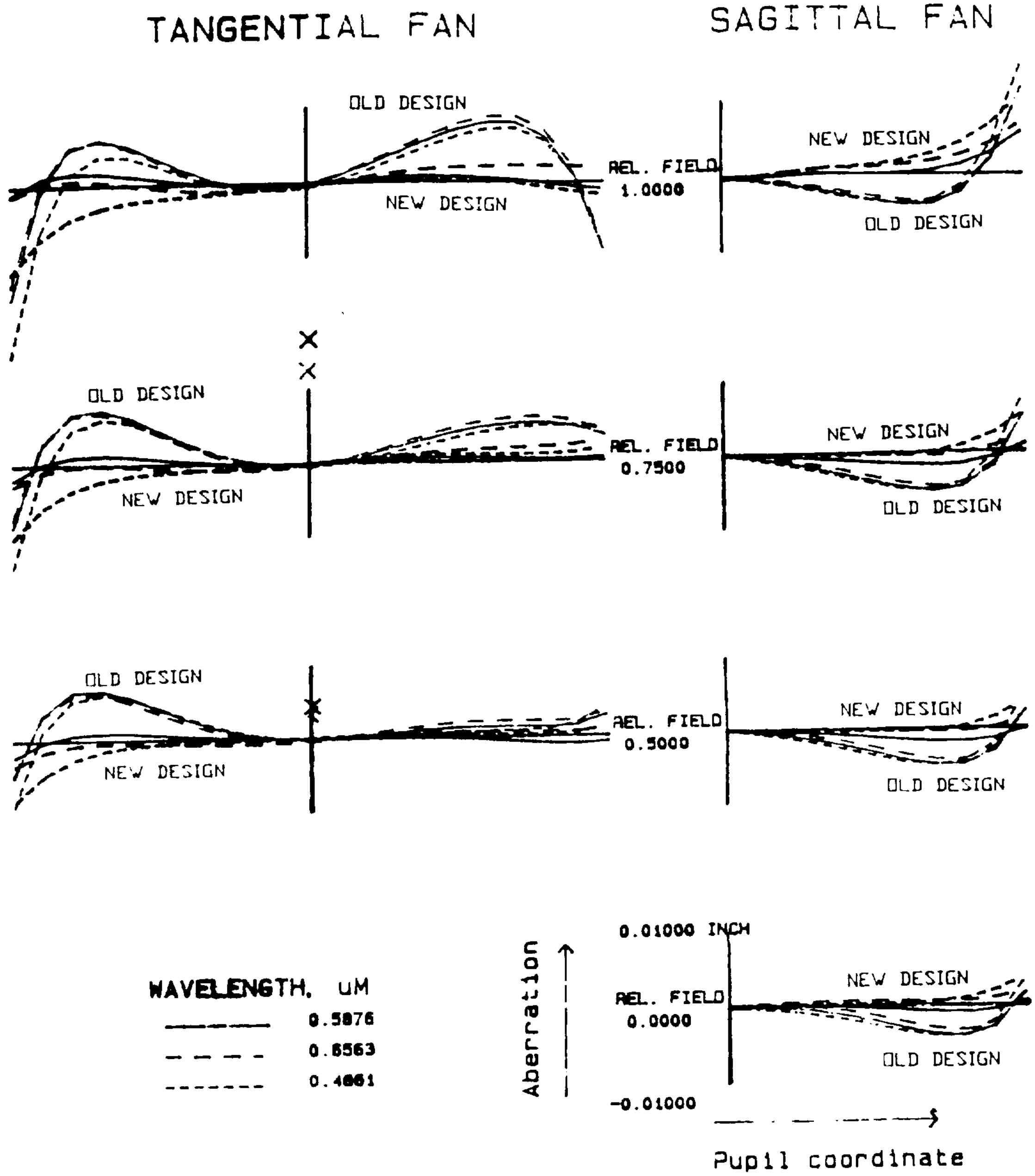


Figure 5. Diagram of Optical Glasses

TRANSVERSE ABERRATION



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SEMI-FIELD = 20.0000 DEGREES SEMI-APERTURE = 1.1111 INCH 10-DEC-89 22: 56: 23

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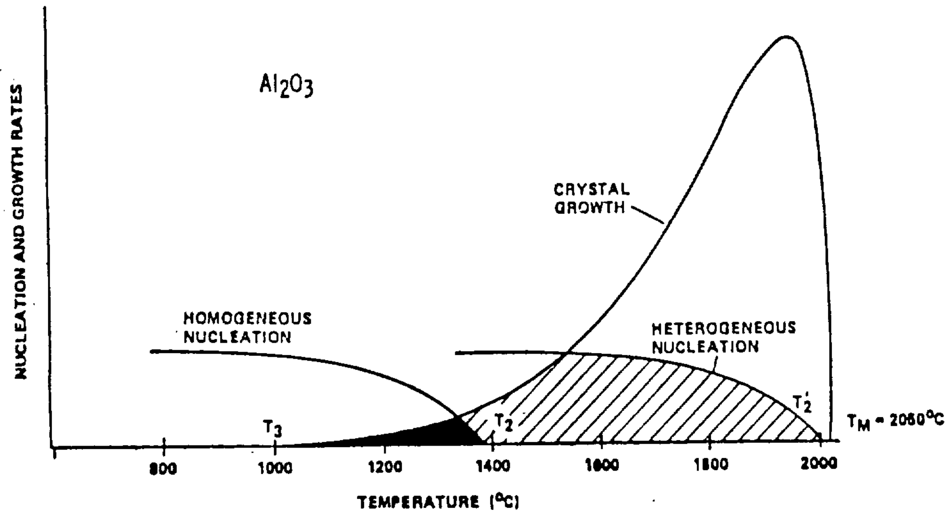
Al2O3
NUCLEATION AND GROWTH RATES
CRYSTAL GROWTH
HOMOGENEOUS NUCLEATION
HETEROGENEOUS NUCLEATION
T3
T2
T'2
TM = 2050°C
800
1000
1200
1400
1600
1800
2000
TEMPERATURE (°C)

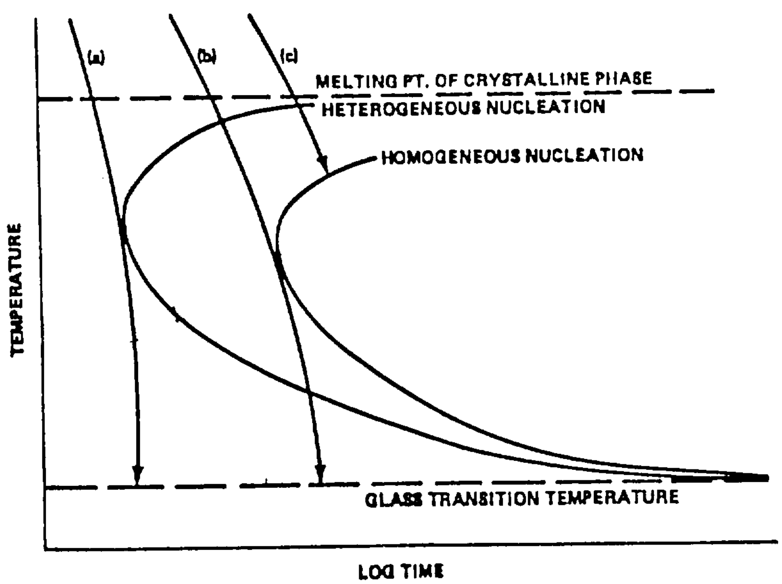
(a)
(b)
(c)
MELTING PT. OF CRYSTALLINE PHASE
HETEROGENEOUS NUCLEATION
HOMOGENEOUS NUCLEATION
TEMPERATURE
GLASS TRANSITION TEMPERATURE
LOG TIME

TABLE I

GROUND-BASED PROCESSING EXPERIMENTS

METHOD	TEMPERATURE LIMIT	COOLING RATE °C/Sec	ADVANTAGES	DISADVANTAGES	EXPERIMENTS
Short Drops Laser Spin Melting	2000°C	1000	Containerless Solidification	No temperature data Very fast quenching Very Small Samples	Refractory rare Earth oxides with alkaline Earth oxides
Laser Drop Melting	2000°C	100 - 500	Large Samples Most oxides	Uncertain quench rate Temperature unknown Large thermal gradients	Ga_2O_3-CaO, Al_2O_3-SiO_2
Long Drop Tube	1800°C	250 - 1000	Containerless Solidification	No temperature data Refractories Difficult	None
Air Jet Levitation/	2000°C	0.1 - 250	Containerless Solidification	Unstable Levitation High Surface Tension	Molten Al_2O_3, Al_2O_3-SiO_2 Glass, Glass Microspheres
Acoustic Levitation/ CO_2 Laser Melting	1000°C	0.1 - 250	Containerless Solidification	Very Unstable Not reproducable	Recrystallized Amorphous Al_2O_3-SiO_2 at 950°C
Thermocouple/ Focused Heating	1750°C	0.1 - 1000	Good Temperature Data	Not containerless Heterogeneous Nucl. Temperature Limits	Ga_2O_3-CaO, Ga_2O_3-CaO-SiO_2, Al_2O_3-CaO

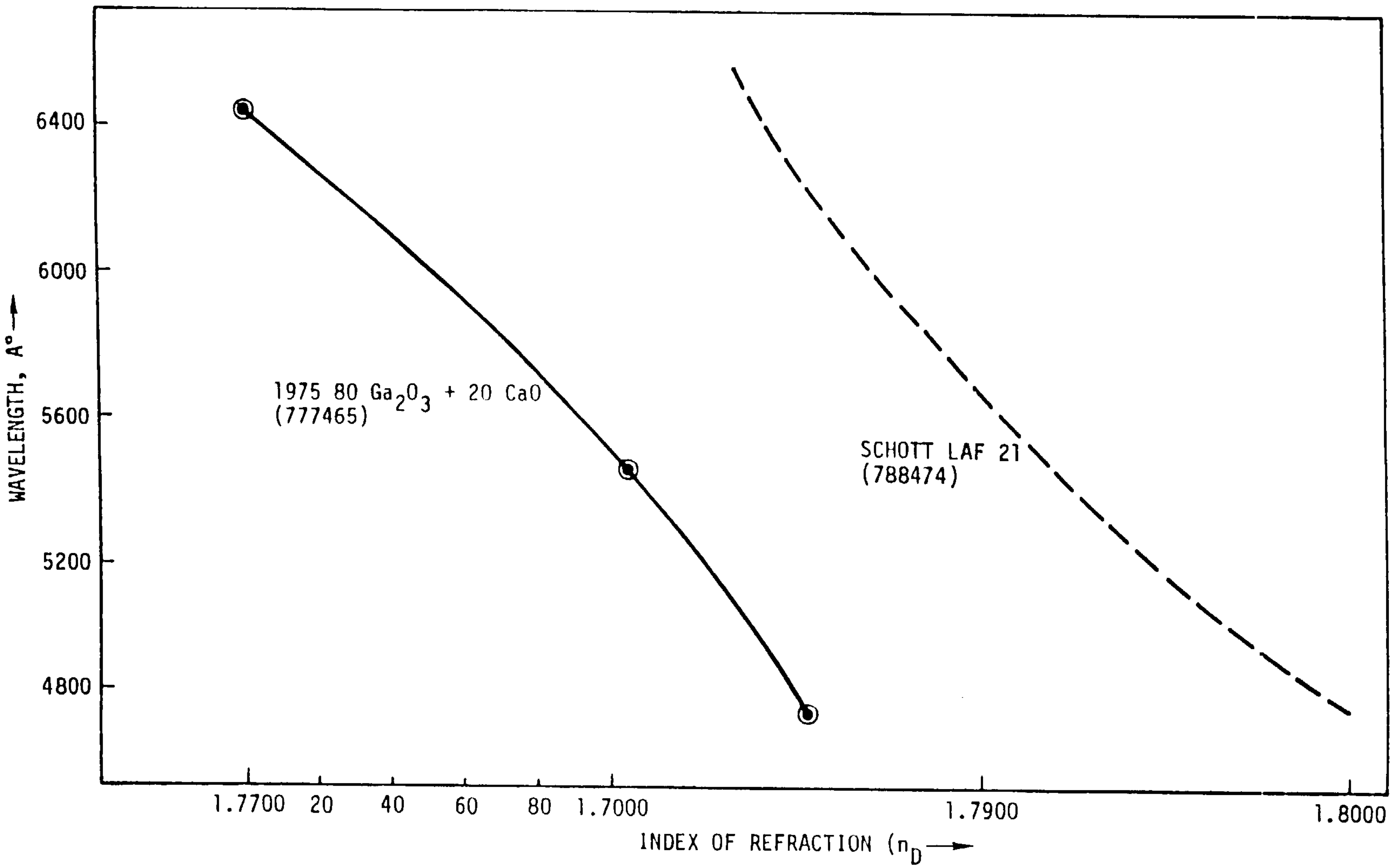

Figure 6. Wavelength Versus Index of Refraction for Ga_2O_3 - CaO Glass and Schott LaF 21 Glass

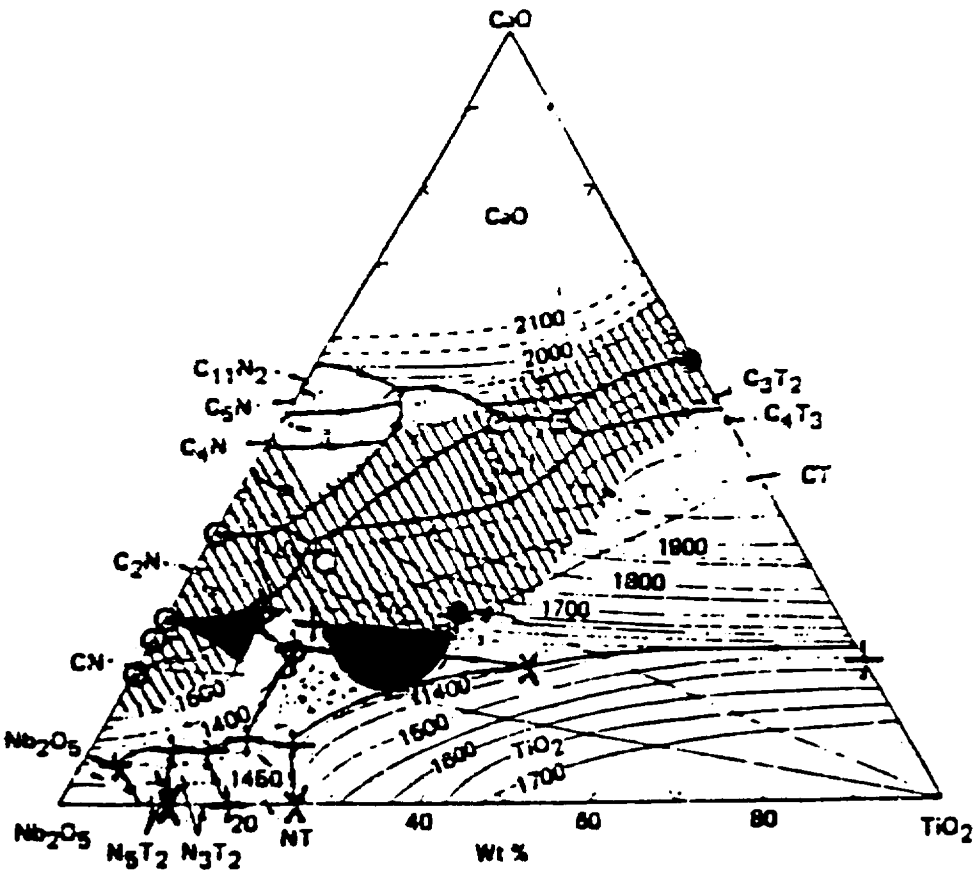

Fig. 5 Glass Formation Region in the Nb_2O_5-TiO_2-CaO System (Phase Diagram in Ref.10) \\\\\

Glass Formation Region in the Above System with $T_m < 1500°C$ ●

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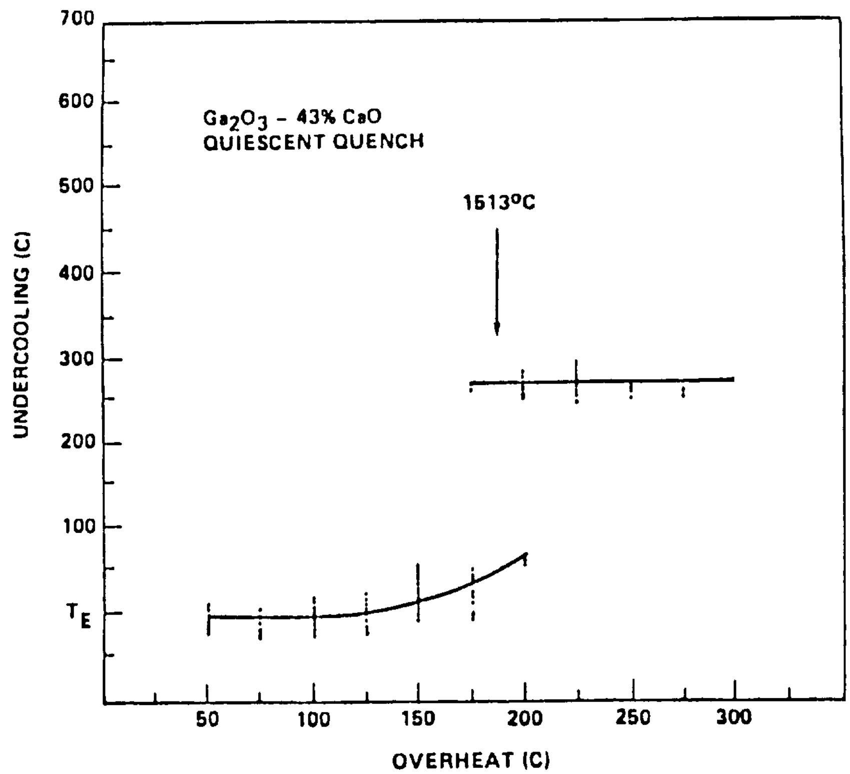

Fig. 4. Plot of undercooling vs overheat for quiescently quenched samples.