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ABSTRACT

The development of laser fusion is highly dependent upon the concurrent development of suitable DT-filled targets. Current programs use hollow glass microspheres as containers for DT gas. The present study was to develop improved materials for the production of these glass microspheres. Results of this study indicate that lithium aluminoborate, aluminosilicate, and borosilicate glasses (in that order of preference) may be used for such applications.

*This work was done in conjunction with Lawrence Livermore Laboratory.

DEVELOPMENT OF GLASSES FOR APPLICATION AS LASER FUSION TARGETS

I. Introduction

The overall development of a practical laser-induced fusion capability is highly dependent upon the development of suitable laser targets. Current programs at Lawrence Livermore Laboratory (LLL), Los Alamos Scientific Laboratory (LASL), and KMS Fusion, Inc. utilize the unique properties of hollow glass microspheres as microcontainers for the mixtures of deuterium and tritium gases which are required for thermonuclear reactions. Glass was selected for this application[1-3] because it can be readily formed into hollow spheres of an appropriate size (40-60 x 10^{-6}m in diameter) which can then be filled with DT gas by a convenient permeation technique[2]. Since most glasses are transparent, optical interference microscopy can be used to characterize[1] the glass spheres as to spheriodicity and uniformity of wall thickness (laser targets must be highly symmetrical.)

Since glass microspheres are an integral part of the laser target, considerable effort is being expended to optimize the properties of this component. Selection of suitable microspheres from commercially available material is a tedious and time consuming task[1] (only one sphere in a million may be usable). Since the commercial material now used is not intended by the manufacturer for such an exacting application, it is believed[4] that considerable improvement in the quality of the microspheres can be achieved. This paper deals specifically with the search for optimum glasses for laser targets.

II. Requirements for Laser Target Glasses

Glasses for the production of laser targets must meet several requirements. First, of course, it must be possible to produce hollow microspheres from such glasses of a suitable quality and in sufficient quantity to meet programmatic needs. However, production of acceptable microspheres would be useless unless the gas permeation properties of the glasses fall within certain limits which result from requirements for the loading and storage

of targets. These limits are, in effect, a compromise between the desire for ease and speed of loading and the need for a reasonable shelf life for the loaded targets. These requirements are currently met[1] by loading the microspheres with high pressure gas (10 MPa) at an elevated temperature (420°C), cooling under pressure, and storing at -30°C. Optimization of this technique would result from the use of glasses having large values of the activation energy for permeation, i.e., permeation coefficients which are very temperature dependent. Further improvement could be obtained from an increase in the loading temperature, since this would allow the use of glasses having a lower permeability at room temperature and below. It follows that very refractory glasses might be desirable for targets if the problems associated with the production of microspheres from such materials can be solved. It is also desirable[4] to have a glass with a low mean atomic number, combined with a minimum in the atomic number of the heaviest element present in the glass, e.g., B_2O_3 is definitely preferable to SiO_2. Since lithium borate glasses have the lowest mean atomic number of the known glass-forming systems,[5] they are obviously the basis for further study. Unfortunately, pure binary lithium borate glasses are not very chemically durable and exhibit a disturbing tendency to react with atmospheric water.[6] This tendency is sharply curtailed by the addition of small quantities of alumina and/or silica to the glass.[6] (It should be noted that the soda lime silicate glass presently supplied by 3M Company is far from ideal - see Table I for composition.)[2]

III. Experimental Procedure

The glasses discussed in this paper were prepared at Sandia Laboratories, Livermore (SLL) from the appropriate oxides (SiO_2, Al_2O_3, B_2O_3) and lithium carbonate by melting in platinum crucibles. All batch chemicals were 99.9% purity or better. Although the glasses have not been analyzed, weight loss during melting measurements indicate that the final compositions are within 1% of those desired.

Gas permeation measurements at SLL have been discussed elsewhere in considerable detail.[7,8] Since the technique was identical to that used in other studies, it will not be repeated at this time.

IV. Results

The present study was intended to be a general survey of gas permeability in low atomic number glasses. Ideally, this study should involve the use of hydrogen isotopes, since the permeabilities[1-3] of interest are those of D_2, T_2, and DT molecules. Unfortunately, the diffusivities of these gases in vitreous materials are very small.[7] Since the total time required for a given permeation measurement is determined by the reciprocal of the diffusivity, it would require a period of weeks per specimen to obtain the desired data, which, of course, would severely limit the variety of glasses studied.

Since a direct measurement of hydrogen isotope permeation appeared to be prohibitively time consuming, it was decided to use helium permeation measurements to identify potential glasses of interest. Once such glasses have been identified, deuterium permeation measurements can be made on a limited number of glasses to define the optimum compositions. This approach is based on the observation that hydrogen isotope permeabilities in commercial glasses are always less than those of helium by two to four orders of magnitude.[2,7] While it has not been shown that this relationship holds for all glasses, the author believes that it is sufficiently valid to be used as a first approximation for a study of this nature.

The temperature dependence of helium permeability in a typical series of glasses is shown in Figure 1. These glasses are described by the compositional formula $X\ Li_2O\cdot(100-X)SiO_2$, where X is the mol% of lithium oxide in the glass. (Values of X for each glass are indicated on the figure.) Data of a similar nature were acquired for each glass studied. These data were extrapolated to 25°C and 300°C and plotted as isothermal permeabilities versus glass composition, e.g., as shown in Figure 2 for some lithium borosilicate glasses. These data were then used to construct the isoperms in each system studied, as shown in Figures 3-8.

Once the isoperms have been determined in a glass-forming system, it is possible to delineate those regions of potential interest for laser targets. If it is assumed that the helium permeability[2] at 300°C of the soda lime silicate glass now used at LLL represents a lower bound at that

temperature, i.e., results in a maximum allowable loading time, and that the value for that glass at 25°C represents a minimum allowable shelf life, it is possible to analyze the data shown in Figures 3-8 to yield regions of potential value. (This assumes that the relative helium/hydrogen permeabilities of the experimental glasses are similar to that of the glass now used.) This has been done for each of the three systems studied to date, as is indicated by the dotted lines in Figures 9-11. Future studies involving hydrogen isotopes can now be concentrated in these regions of glass formation.

V. Discussion

Although each of the three ternary glass-forming systems studied to date appear to exhibit regions of interest for laser targets, other factors must be considered in choosing the most appropriate for additional, detailed study. Characteristics such as chemical durability, tendency toward phase separation and/or crystallization, and maximum loading temperature also are quite important in the selection of target glasses. The lithium borosilicate glasses, for example, appear to present considerable problems due to their tendency to phase separate, i.e., to separate into two distinctly different glasses. The physical appearance of each of the glasses studied is indicated in Figure 12. The dashed lines represent the approximate areas of phase separation in this system. Although many of these glasses exhibit acceptable permeation characteristics, it must be emphasized that gas permeation in such glasses is strongly influenced by morphology,[9] which, in turn, is determined by the thermal history of the glass.[10] Since the process for manufacturing hollow microspheres results in a radically different thermal history from that of the bulk specimens of the present study, it may be very difficult to extrapolate these results to practical values for microspheres.

The other two systems studied also exhibit potential problems. Lithium aluminosilicate glasses are known for their outstanding chemical durability and high use temperatures, but do display a disturbing tendency to crystallize at elevated temperatures. The lithium aluminoborate glasses have the lowest mean and maximum atomic numbers of the glasses studied, but have relatively low maximum use temperature limits. Furthermore, while bulk

specimens appear to be quite durable if they contain at least a few mol% alumina,[6] essentially nothing is known about the long-term durability of these glasses.

VI. Conclusion and Recommendations

All three of the glass-forming systems studied to date exhibit permeability regions which would be satisfactory for laser targets. However, other considerations (see Section V) lead the author to recommend the lithium aluminoborate and aluminosilicate glasses (in that order) over the lithium borosilicate glasses. If other criteria, such as manufacturability, favor the latter, additional study of lithium borosilicate glasses containing a few percent alumina would be quite valuable, since such glasses might well be single phase. (Alumina is known to suppress phase separation in other borosilicate systems.)[10]

The glass-forming systems studied to date have all contained lithium oxide. However, SL studies[6,9] of similar systems containing other alkali oxides have indicated that much more desirable glasses (at least insofar as permeation is concerned) could be made in the corresponding sodium oxide systems. This would, of course, increase the mean atomic number of the glass. However, since only a maximum of 30 to 40 mol% of the composition would be alkali oxide, this penalty is not as great as might be imagined. A comparison of the mean atomic number of some suggested glasses is shown in Table II. Since the sodium base glasses also may have a number of other advantages (better chemical durability, less tendency to phase separate and/or crystallize, probably better viscosity versus temperature relationships), we would strongly suggest that some consideration be given to additional study of those glasses.

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Table I

Composition of Soda-Lime-Silica Glass Microspheres

Component	Mol%
SiO_2	79.0
CaO	10.0
Na_2O	6.8
B_2O_3	2.0
ZnO	0.8
MgO	0.5
P_2O_5	0.3
Al_2O_3	0.2
K_2O	0.1
Others	0.3

Mean atomic number = 10.4

Maximum atomic number = 30

Atom % > Si = 3.8

Table II

Mean and Maximum Atomic Number of
Some Simple Tenary Glasses

Glass Composition	Mean Atomic Number	Maximum Atomic Number
$Li_2O \cdot B_2O_3 \cdot SiO_2$	7.15	14
$Na_2O \cdot B_2O_3 \cdot SiO_2$	8.93	14
$Li_2O \cdot Al_2O_3 \cdot B_2O_3$	7.15	13
$Na_2O \cdot Al_2O_3 \cdot B_2O_3$	8.93	13
$Li_2O \cdot Al_2O_3 \cdot SiO_2$	8.22	14
$Na_2O \cdot Al_2O_3 \cdot SiO_2$	10.00	14

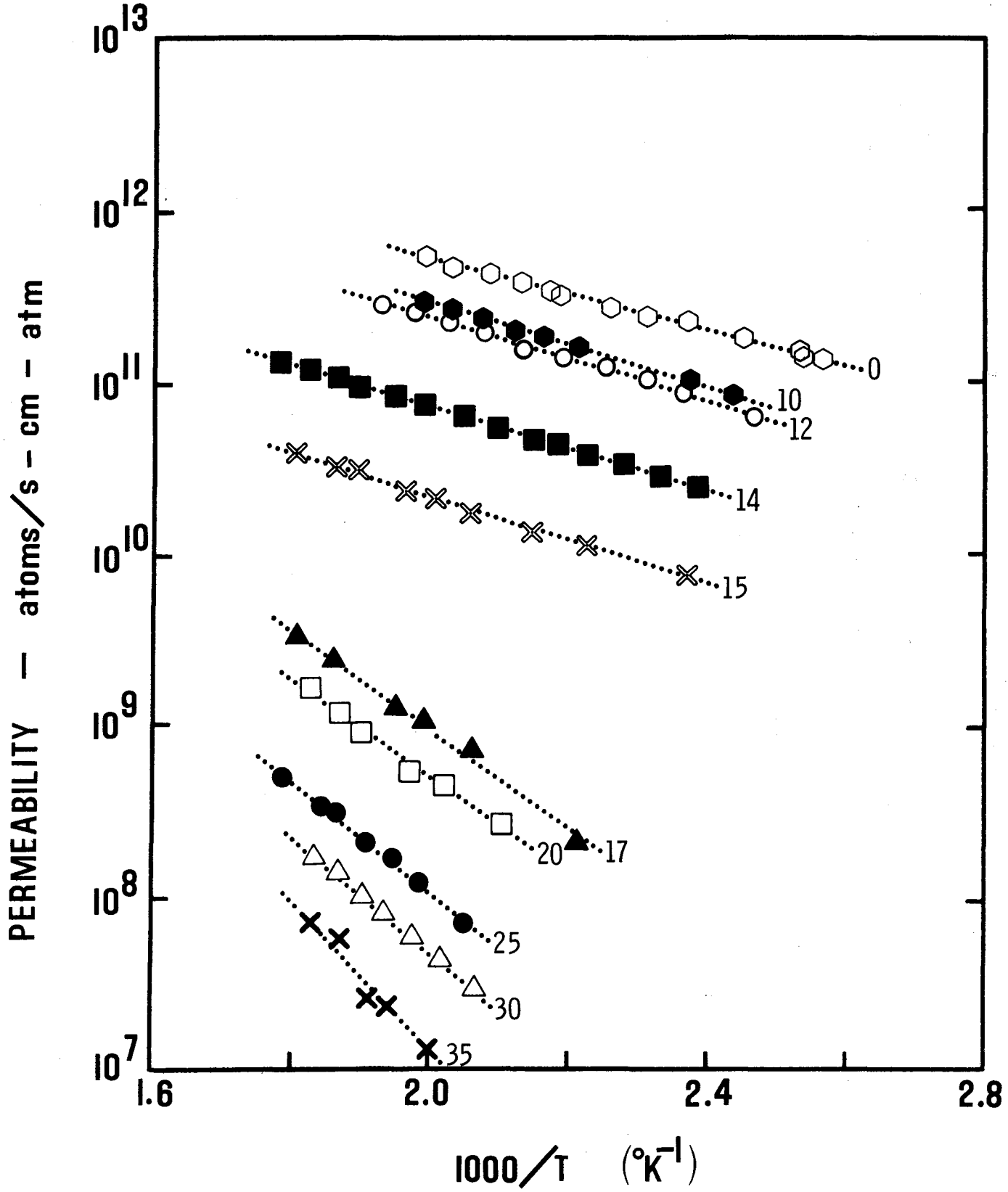

Figure 1: Temperature dependence of helium permeability in lithium silicate glasses of the general formula X $Li_2O \cdot (100\text{-}X)SiO_2$. The value of X for each glass is indicated on the figure.

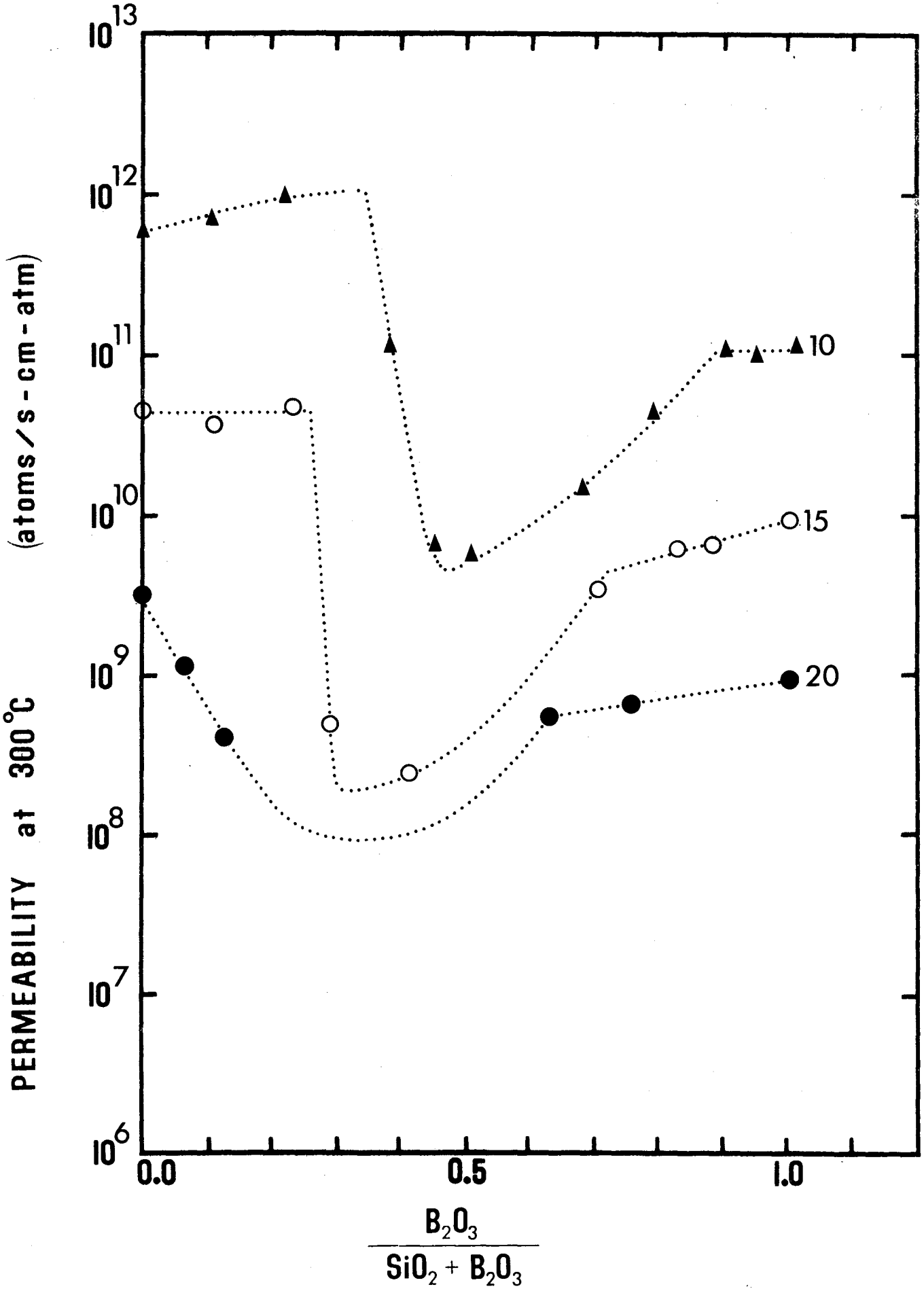

Figure 2: Isothermal permeabilities of some lithium borosilicate glasses. The mol% Li_2O for each series of glasses is indicated on the figure.

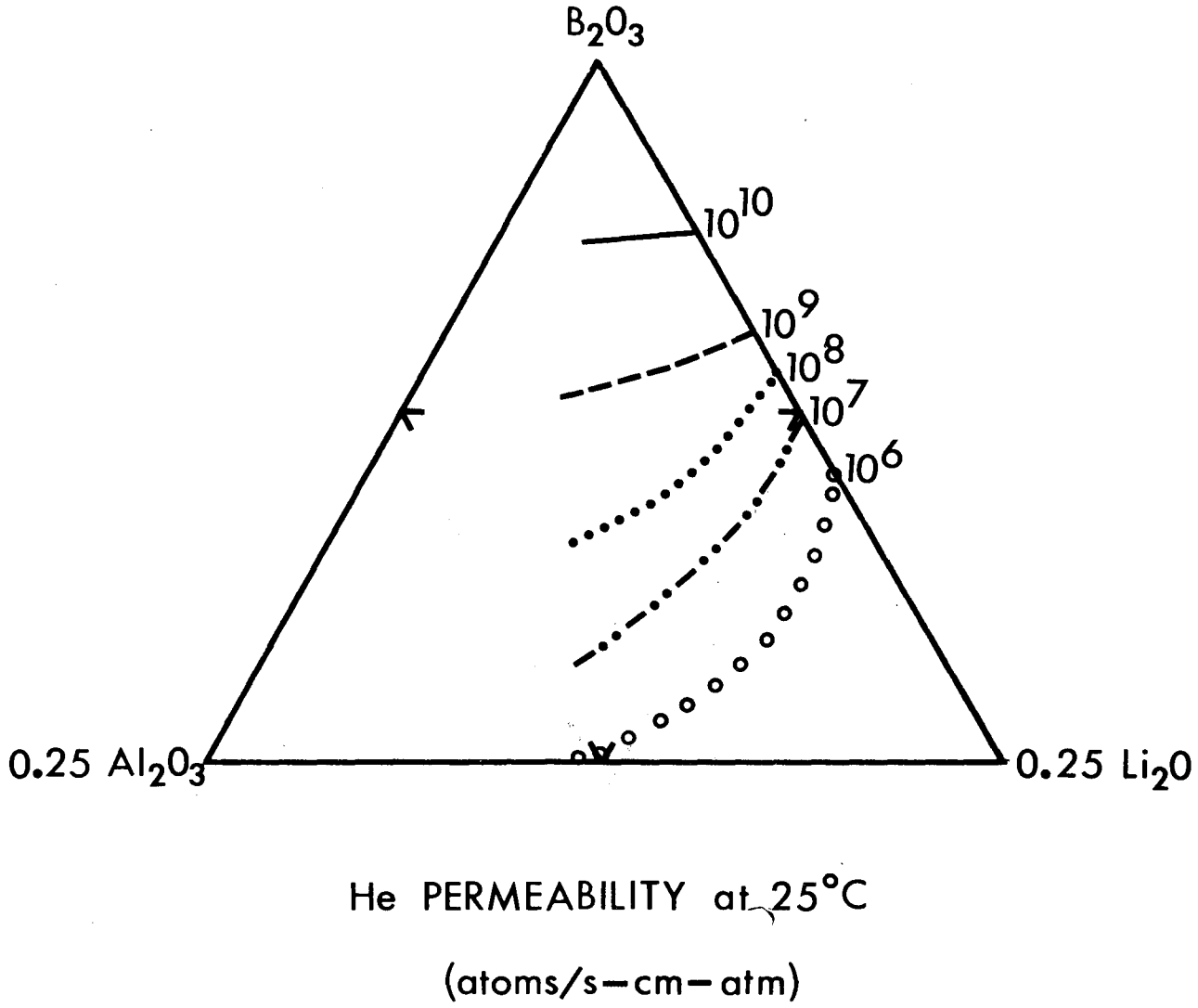

Figure 3: Isopermeation lines at 25°C for lithium aluminoborate glasses.

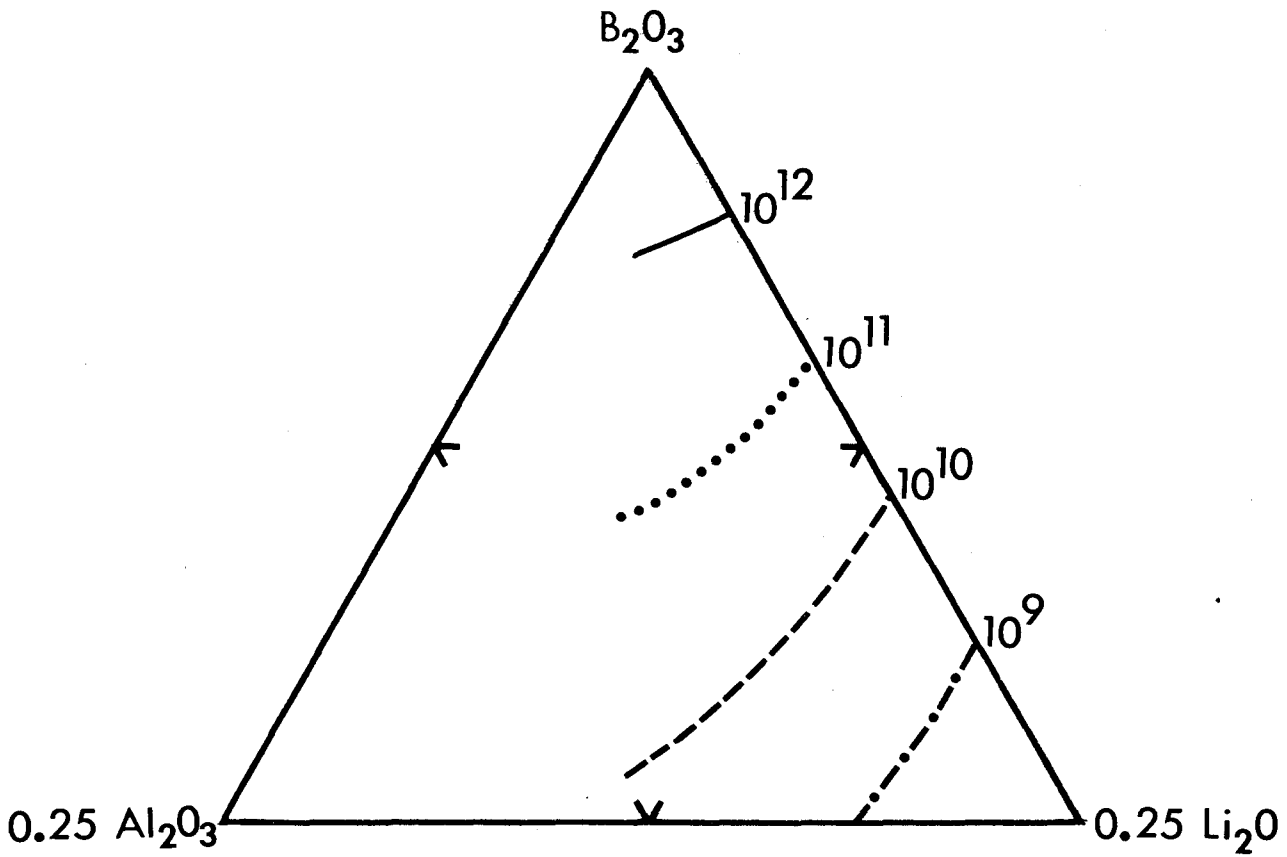

Figure 4: Isopermeation lines at 300°C for lithium aluminoborate glasses.

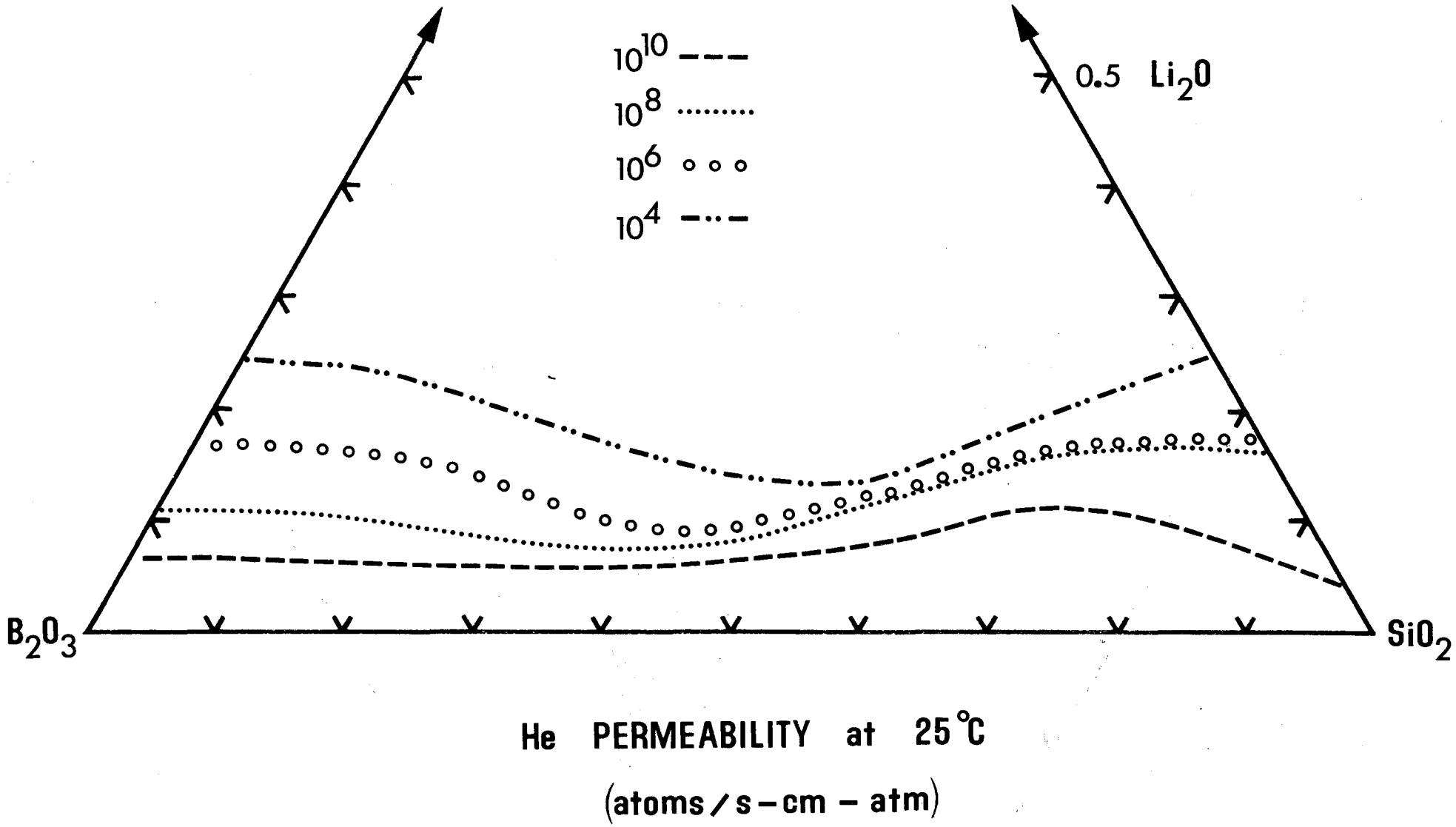

Figure 5: Isopermeation lines at 25°C for lithium borosilicate glasses.

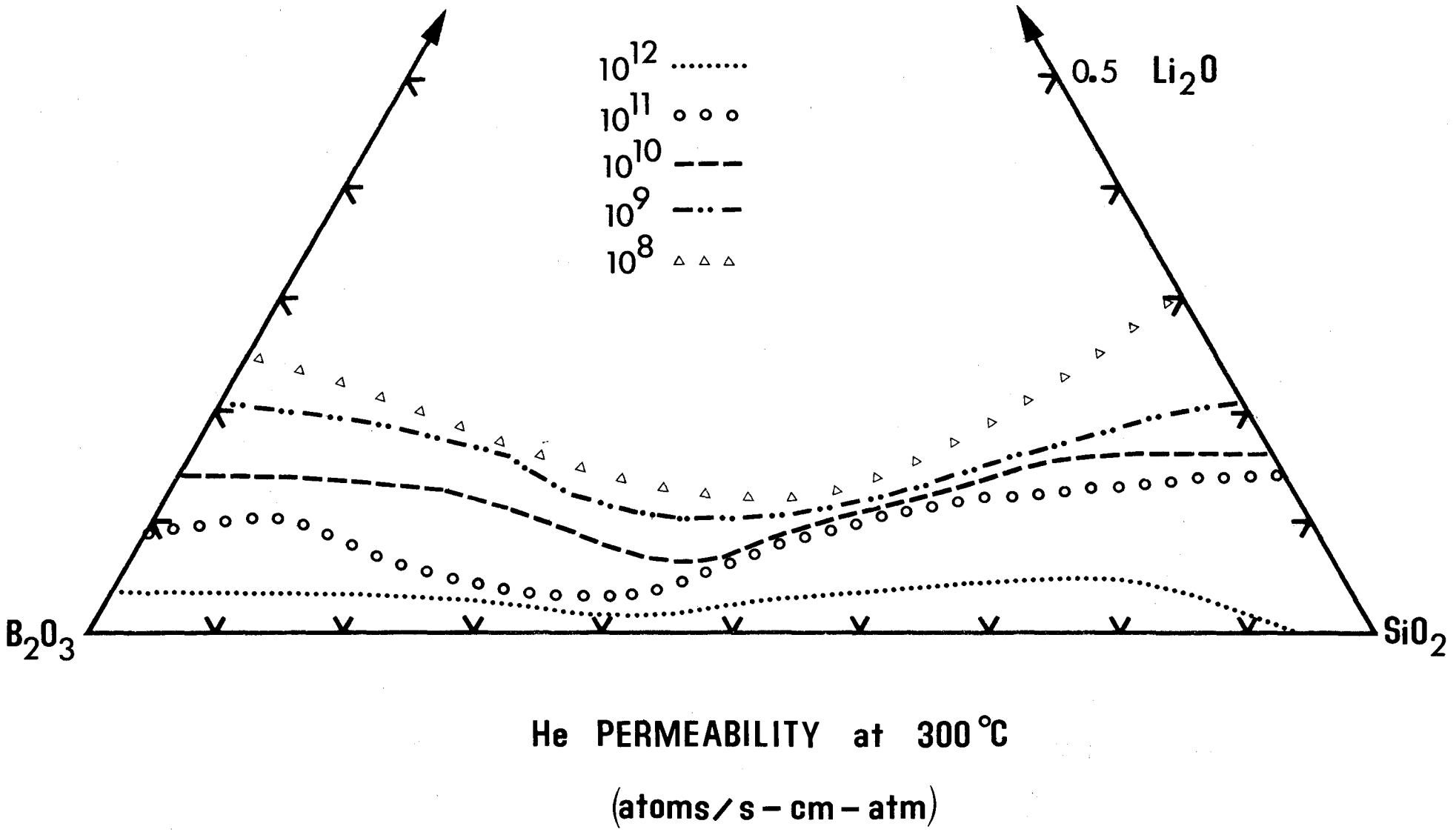

Figure 6: Isopermeation lines at 300°C for lithium borosilicate glasses.

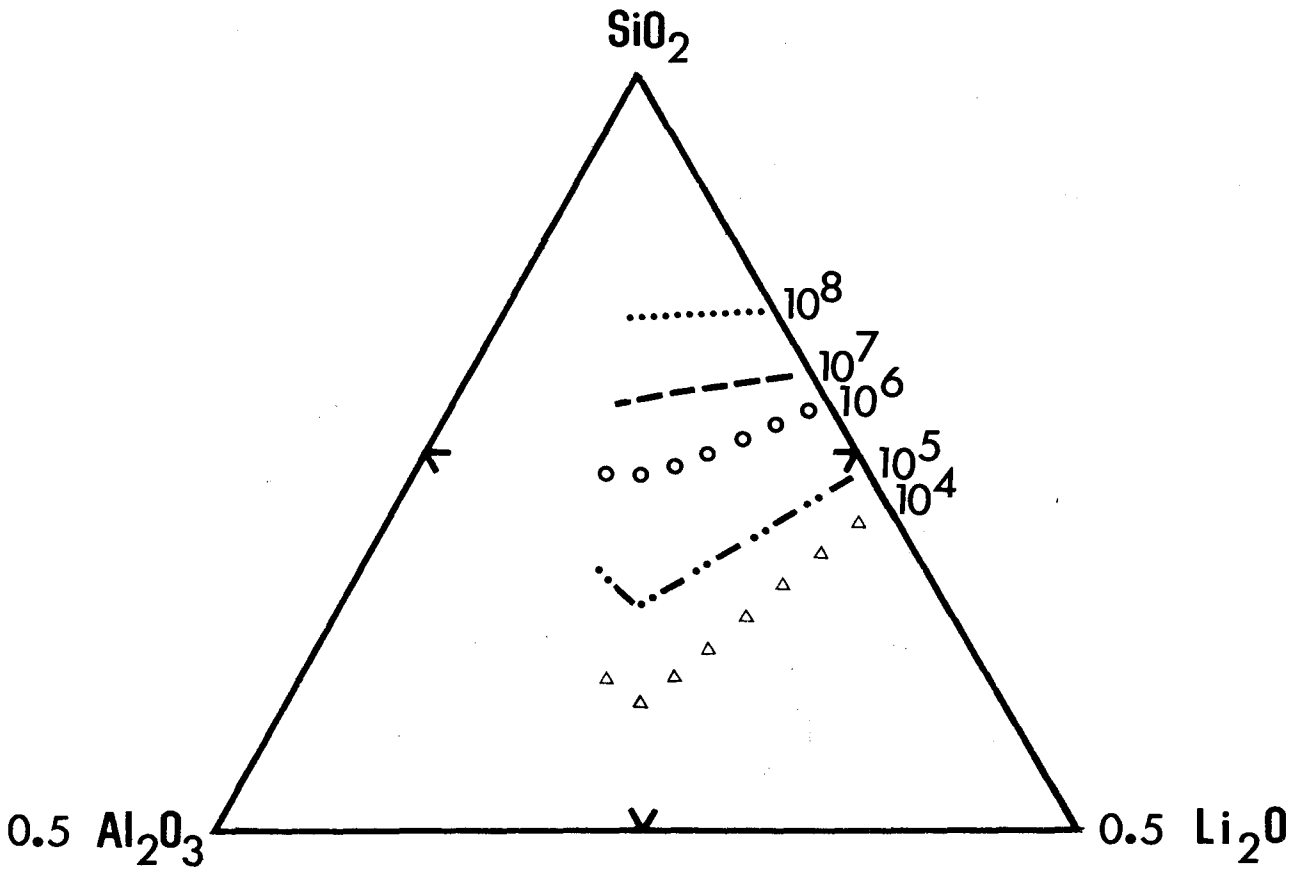

Figure 7: Isopermeation lines at 25°C for lithium aluminosilicate glasses.

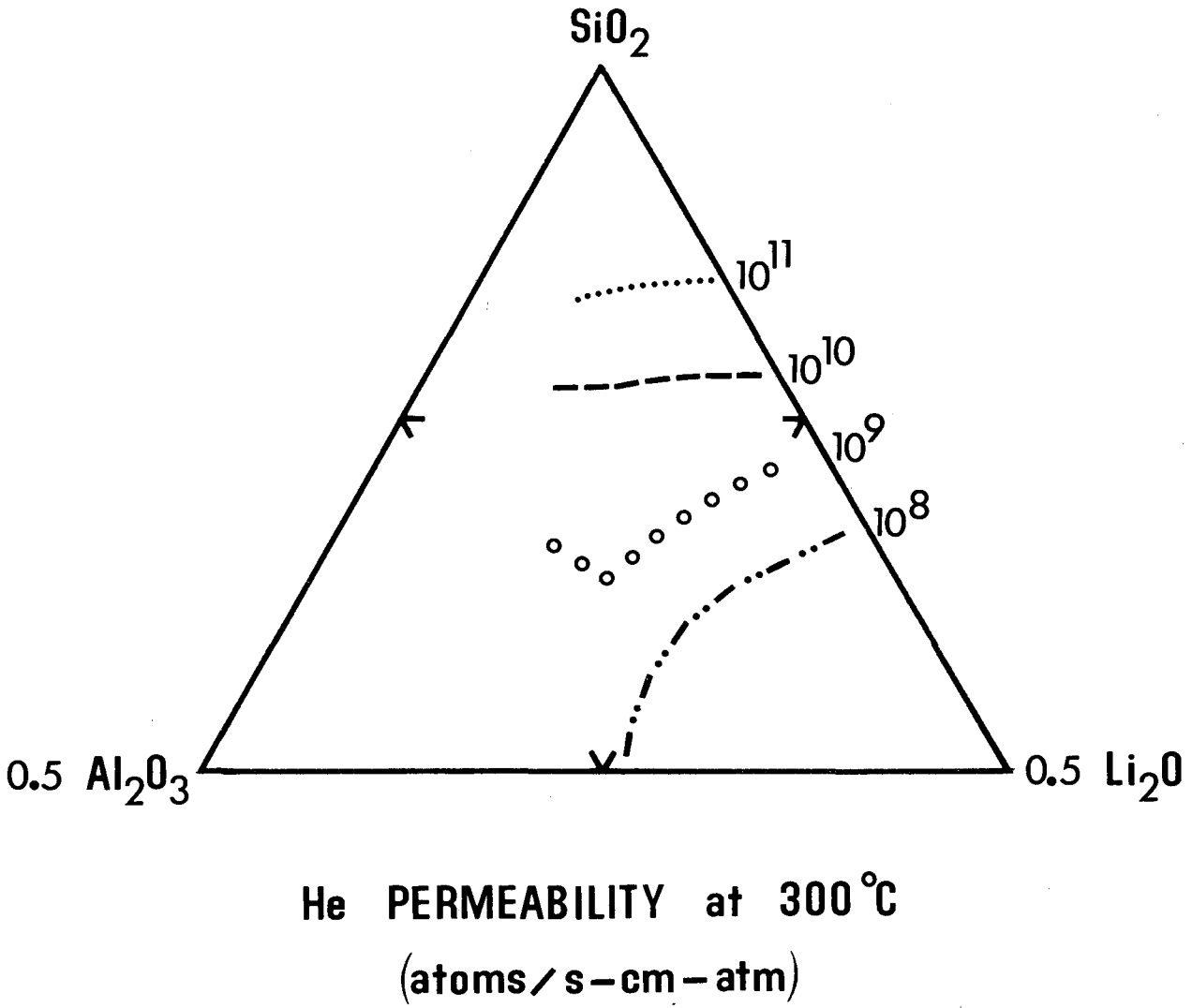

Figure 8: Isopermeation lines at 300°C for lithium aluminosilicate glasses.

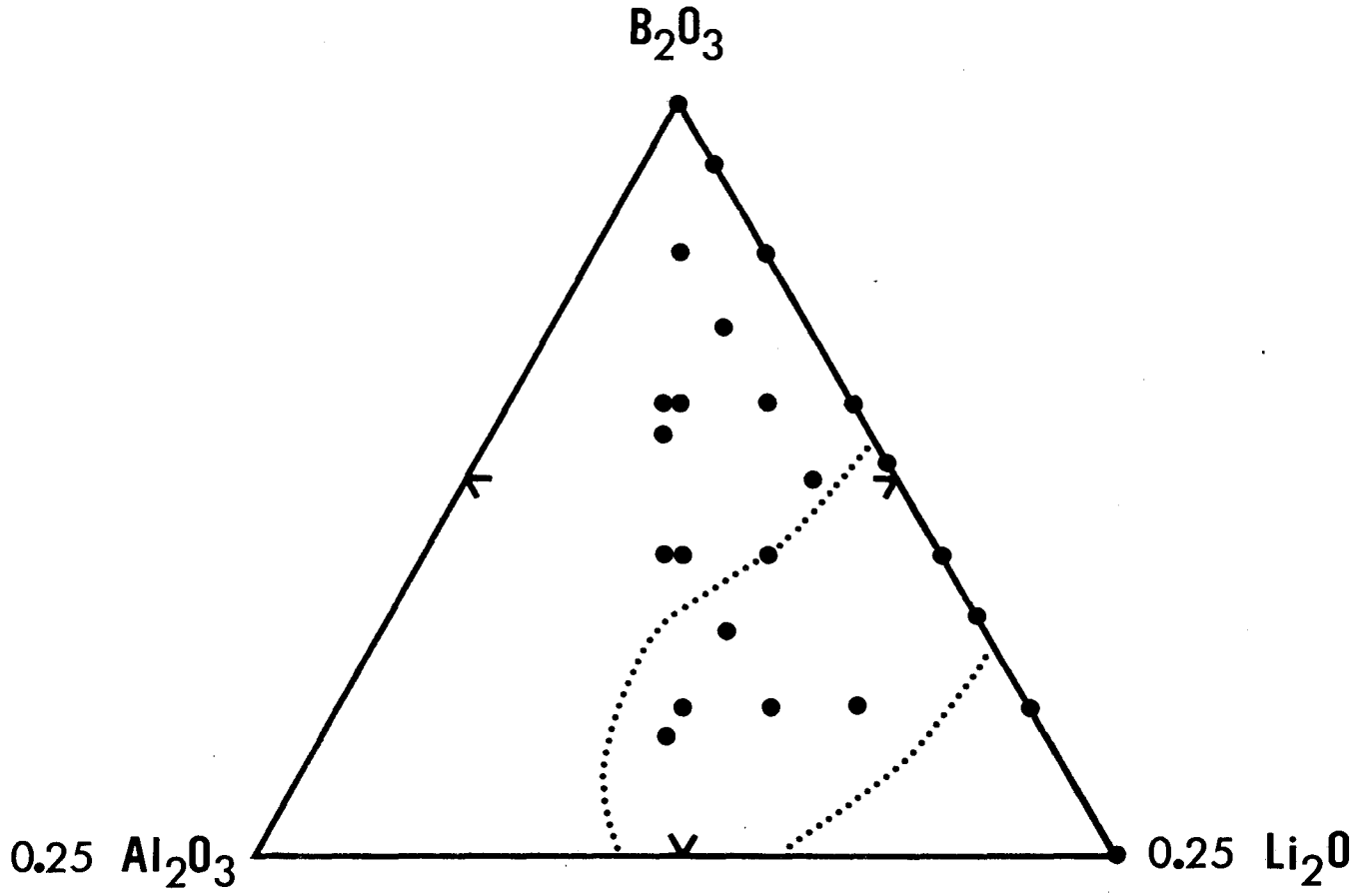

Figure 9: Region of potential laser target glasses in the lithium aluminoborate system (indicated by the dotted line). Solid circles indicate compositions studied.

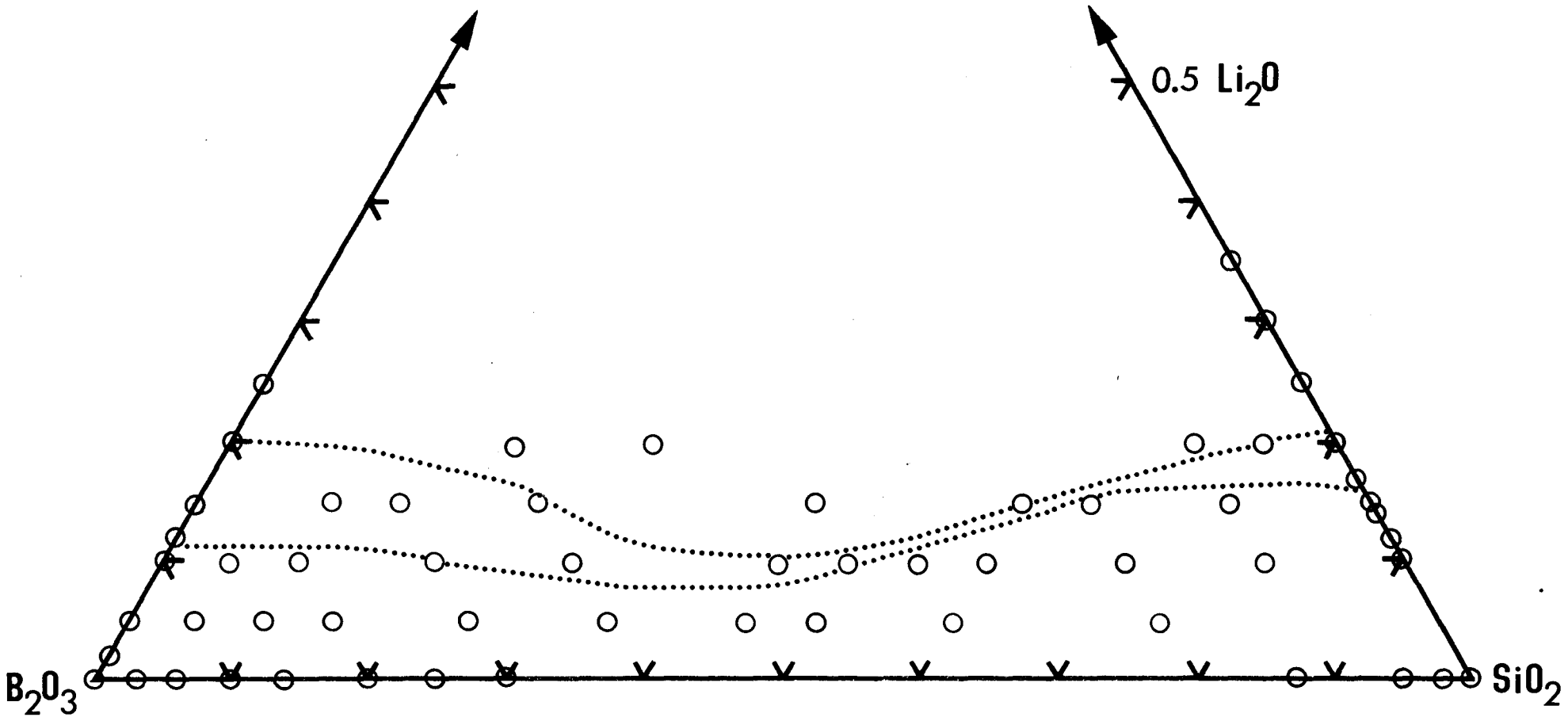

Figure 10: Region of potential laser target glasses in the lithium borosilicate system (indicated by the dotted line). Open circles indicate compositions studied.

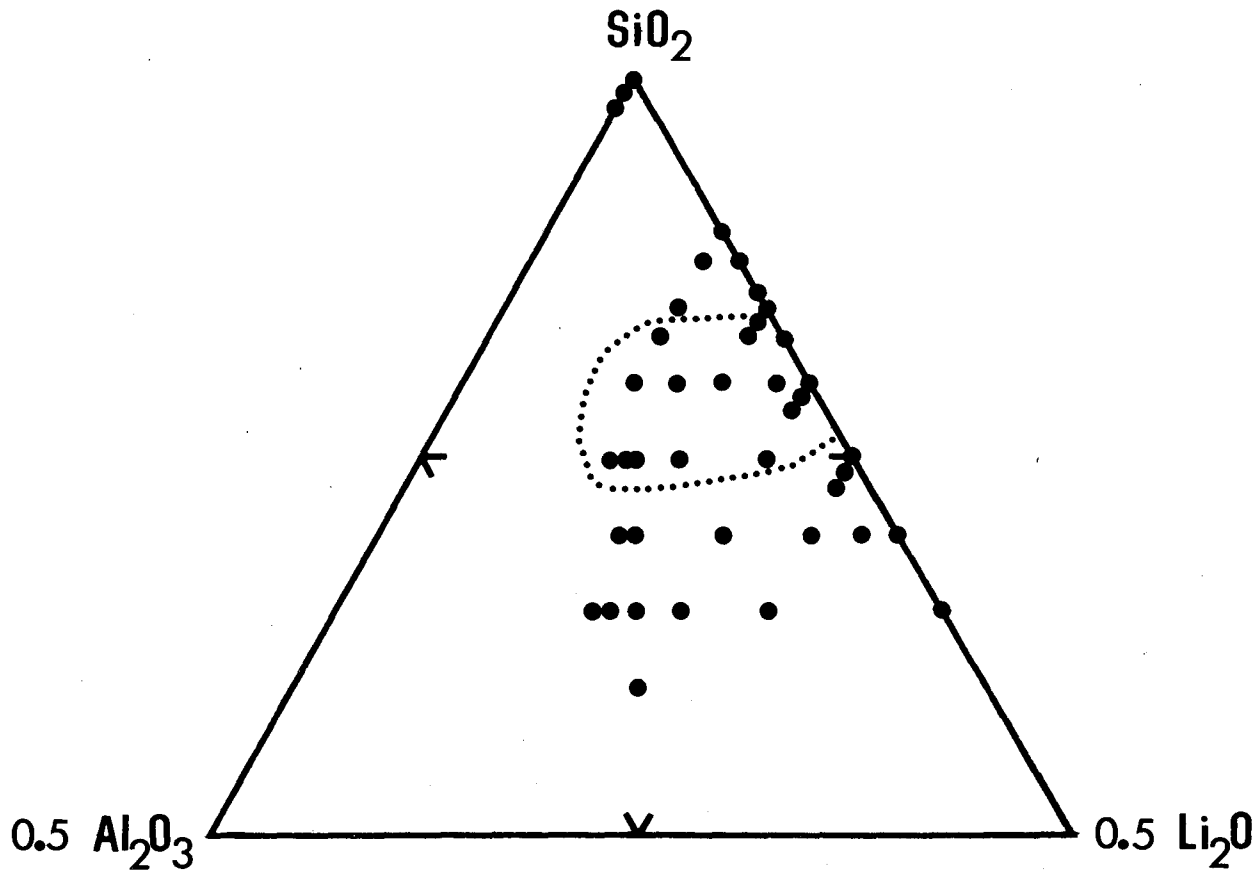

Figure 11. Region of potential laser target glasses in the lithium aluminosilicate system (indicated by the dotted line). Solid circles indicate compositions studied.

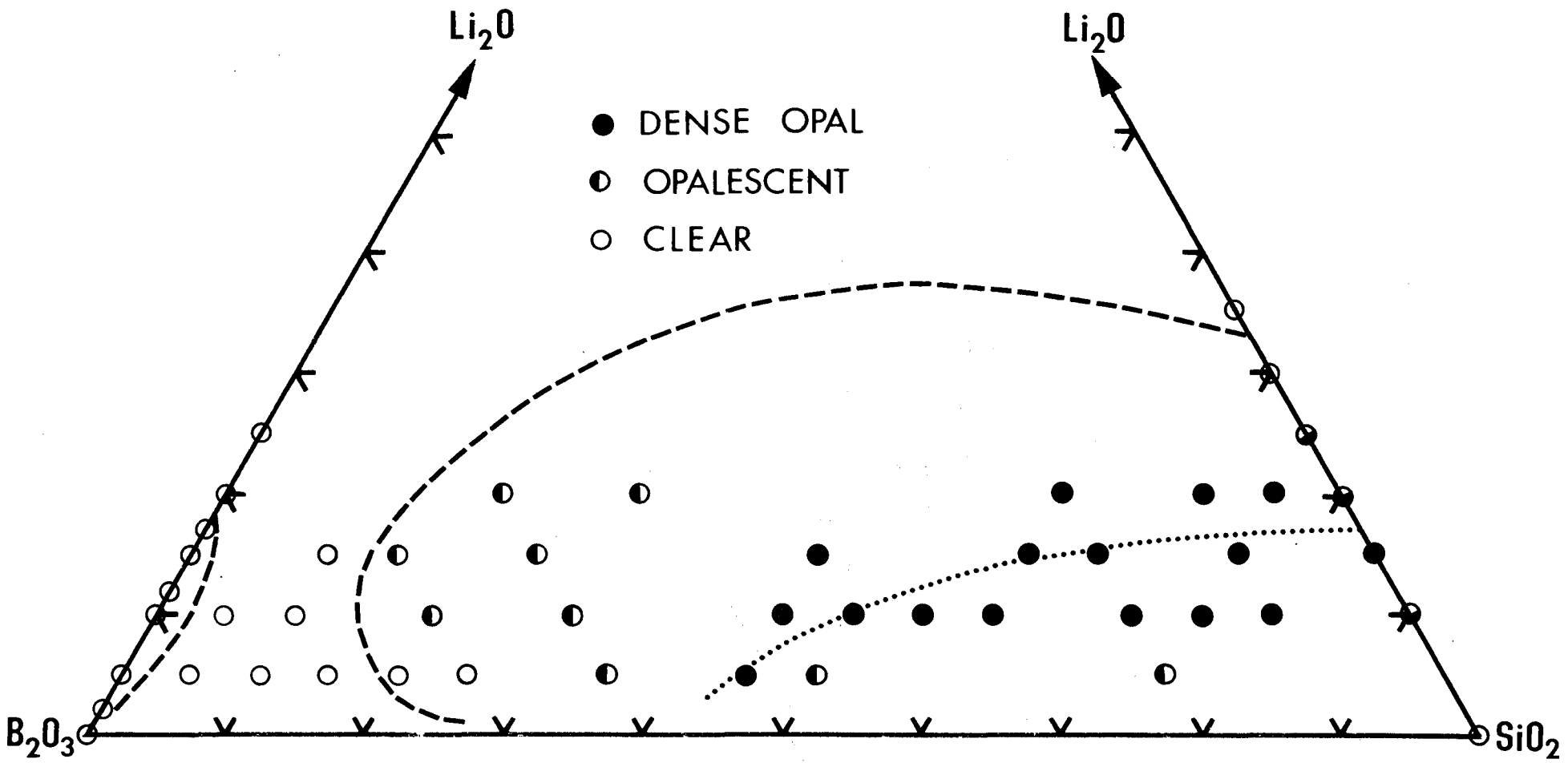

Figure 12: Regions of phase separation (dashed lines) in the lithium borosilicate system. The physical appearance of each glass is indicated on the figure.

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