REFRACTIVE INDEX OF CaAl₂Si₂O₈ GLASS AND CRYSTAL ANORTHITE AT PRESSURES UP TO 5.0 GPA

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Introduction. The pressure behaviour of the refractive index of $CaAl_2Si_2O_8$ glass (d=2.69 g/cm³) was studied in a range 0-5.0 GPa. This glass was chosen as the object of investigation because of 1) the glass composition corresponds to those of the mineral anorthite (the end-member feldspars), 2) as contrasted to others glasses of the end-members feldspars the structure of $CaAl_2Si_2O_8$ glass is similar with the crystal structure [1].

Experimental technique and results. $CaAl_2Si_2O_8$ glass was prepared by melting of the component oxides in Pt crucibles at 1550 °C for 1 hr, remelting and quenching in water. No annealing step was made.

The refractive indices at atmospheric and high hydrostatic pressures were measured according to the developed technique described in detail in works [2, 3] with the use a polarizing interference microscope and diamond anvil cell. Polished glass plates 15-20 μ m in thickness were prepared for investigation. The optical path difference Φ and the refractive indices of the sample n_x and a surrounding medium n are related by the expression $\Phi=(n_x - n)/t$. The method for measuring is based on the pressure- induced change in the optical path difference Φ caused by the sample. It can be seen from the expression that a decrease in the optical path difference Φ results from the change in the ratio between the refractive indices of the sample n_x and the surrounding medium n (under pressure , the refractive index of the liquid increases considerably faster than the refractive index of the glass). The error in the determination of the pressure and the refractive index were respectively $\delta P=\pm 0.05$ GPa and $\delta n_x =\pm 0.003$. The pressure dependence of the refractive index for crystal anorthite studied earlier in [4].



Fig. Dependences of the refractive index for glass $CaAl_2Si_2O_8$ and crystal anorthite n on the pressure *P*: rhombus - $CaAl_2Si_2O_8$ glass, square– crystal anorthite [4].

Discussion. At the range pressures 0-0.1 GPa the refractive index of glass increases abruptly up to n=1.596. This behaviour is not typical for glasses. At the ranges pressures 0.1-1.5 GPa and 1.5-4.5 GPa the refractive index of the glass increases with a baric coefficient $\Delta n/\Delta P=0.012$ GPa⁻¹ and $\Delta n/\Delta P=0.005$ GPa⁻¹ respectively. The pressure dependence of the refractive index for the crystal anorthite has an anomaly in the range 2.6-2.9 GPa.

The crystal structure of anorthite has been determined by X-ray diffraction method [5]. Anorthite is a triclinic crystal belonging to the plagioclase series. High - pressure study revealed the polymorphous transformation in the range 2.6-2.9 GPa, which is attended by a small volume change of a unit cell (0.2%). In the high-pressure structure the internal dimensions of the tetrahedral as represented by average T-O distances and O-T-O bond angles show no significant changes from the low-pressure structure. By contrast, changes in T-O-T bond angles are observed and Ca-O bond lengths are appreciably shortened in the high-pressure structure. The mechanism for the transition is the tilting of essentially rigid tetrahedra, apparently driven by a

need to accommodate both the applied pressure and the large Ca cations with the cavities of the framework. The anomaly, which shows the pressure dependence of the refractive index of the crystal anorthite, caused by the phase transition in the range 2.6-2.9 GPa.

The structure of $CaAl_2Si_2O_8$ glass has been determined by different methods. A brief overview is reviewed in [6]. The investigations show that the $CaAl_2Si_2O_8$ glass is similar in the structure with the crystal anorthite. The structure has SiO_4 and AlO_4 tetrahedra which are connected to other four – membered rings in a more or less random fashion. The initial abrupt increasing of the refractive index of $CaAl_2Si_2O_8$ glass, Fig., can be clarified by the structural similarity of the glass and crystal anorthite and by the purpose of the system to achieve more tolerant state. In the following increasing of the pressure at the range 0.1-1.5 GPa the baric coefficient of the refractive index of the glass is yigher than this for the crystal anorthite and at the range 1.5-4.5 GPa the baric coefficient is in a close agreement with those for the crystal anorthite at the pressures range after phase transition.

In spite of the structural similarity there are distinctions between the behaviour of the glass and crystal anorthite at high pressures. Crystals compress through changes in bond angles and distances that are consistent with the crystal structure. These changes are termed vibrational since they involve only displacement of atoms within the potential energy wells of the structure [7]. The main reason for the greater compressibility of the glass and melt is the existence of a configurational contribution to the compressibility in addition to vibrational one. The configurational component ensures change of the glass structure up to the coordination changes under the pressure. However, even these changes in the glass occur continuously as contrasted to the crystal. It is not to be expected an abrupt anomaly in the pressure dependence of the refractive index (n) for the CaAl₂Si₂O₈ glass.

According to the data available to us now it is impossible to judge the structural changes taking place in glass in the range pressures 0-5.0 GPa. It is reasonably safe to suggest that in the pressures range 1.8-4.5 GPa the $CaAl_2Si_2O_8$ glass structure is similar with the high - pressure structure of anorthite.

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