Neutron Diffraction from Amorphous Quartz

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Abstract : The structure of amorphous quartz (SiO_2) has been investigated in the neutron diffraction method. The structure factor S(Q), atom pair correlation function g(r) and radial distribution function Rh (r) have been determined. The distances between the two Si-O, one Si-Si and two O-O atom pairs have been determined through this investigation and compared with those of reported values.

Keywords : *Neutron diffraction, Quartz, Atomic distances, Amorphous materials.*

INTRODUCTION

The atomic structure of liquid and amorphous materials are intermediate between the perfect disorder of atoms or molecules in a gas and the highly perfect three dimensional order in a solid. The immediate environment of an atom in an amorphous material is not very different from that in solid [1]. It may be thought that a quartz or glass is a supercooled liquid or as a liquid with extremely high viscosity which, with change of temperature, falls rapidly as the softening point is approached. When the structure of glass is investigated by neutron diffraction studies, it is the similarities and the differences between a glass and the extremes of solid and liquid which are important. In an amorphous material, as in a liquid, there is short-range atomic order but no long-range order. On the other hand, in quartz, as in a solid, the individual atoms have defined equilibrium positions, and they are not able to diffuse through the body of the material. The study of amorphous solids is important for understanding the state of matter that is intermediate between the crystalline and the liquid states [2]. Predictions and explanations of the formation, of amorphous solids are to date incomplete and semi-empirical [2]. For glass formation, for example, there are several kinetic structural theories [3] that have predicted formation with varying degree of success. Oxide glasses, in particular, often conform to a set of simple rules proposed by Zachariesen [4] and Crystallite theories [5] differ in

Table 1 : Atomic Distances for Amorphous Quartz Sample

Si-O ($\overset{o}{A}$)	O-O ($\overset{o}{A}$)	Si-Si ($\overset{o}{A}$)	Si-O ($\overset{o}{A}$)	O-O ($\overset{o}{A}$)	Reference
1.60	2.64	3.2	4.0	5.0	10
1.58	2.56	3.02	4.0	5.1	11
1.61	2.62	3.1	4.1	4.98	12
1.62	2.61	3.2	4.1	5.0	present study

extent to which there are fluctuations in local order. At present a number of different theories based on a wide variety of concepts e.g. topology, ring statistics, etc. are being considered. The success of any such theory will of course depend on agreement with macroscopic as well as microscopic experimental data e.g. diffraction data.

The oxide glass SiO_2 is considered to be the sturctural analogue of vitreous $ZnCl_2$ together with BeF_2 [6]. The neutron diffraction technique has emerged as an important tool in studying the structure of amorphous materials [7,8]. The dynamical structure factors of different amorphous materials are also being studied [9,10]. The newly set-up triple axis spectrometer at the Atomic Energy Research Establishment (AERE), Savar, Dhaka has been employed to study the amorphous solid, in this case, a quartz sample by neutron diffraction technique. The result obtained from the present measurement has been compared with those cited in literature [10, 11, 12]. The agreement between the g(r) functions of present work and the work reported in reference [12] is quite good.

EXPERIMENT

Cylindrical quartz tube has been used as the sample. Measurements have been carried out with a neutron beam of wavelength 1.24 $\overset{o}{A}$. The double axis mode of

the triple axis spectrometer (TAS) at the piercing beam port of the TRIGA Mark II research reactor of AERE, Savar has been employed for this experiment. BF3 detector and monitor were used for this measurement. The measurements were carried out covering angles with values of 2θ, 10 - 90° in steps of 0.2°. The whole sample is bathed in the neutron beam. The backgrounds at initial angles were corrected properly. The data were analyzed using a computer program developed for this purpose.

THEORY

The differential scattering cross-section is given by [1]

$$\frac{d\sigma_{coh}}{d\Omega} = Nb^2 \left[1 + \frac{4\pi\rho}{Q} \right] \int_0^\infty \{g(r)-1\}\, r \sin Qr\, dr \ldots\ldots \quad (1)$$

where Q is the wave vector transfer, N is the number of atoms per unit volume, g(r) is the pair correlation function, b is the neutron scattering length and ρ is number density of the atoms .

The structure factor S(Q) is related to coherent scattering cross-section as

$$\frac{d\sigma_{coh}}{d\Omega} = Nb^2\, S(Q) \ldots\ldots \quad (2)$$

From here we may write an expression for structure factor S(Q) as

$$S(Q) = 1 + \frac{4\pi\rho}{Q} \int_0^\infty \{g(r)-1\}\, r \sin Qr\, dr \ldots\ldots \quad (3)$$

From S(Q) the pair correlation function can be obtained by Fourier transformation

$$g(r) = 1 + 1 + \frac{1}{nr} \int_0^\infty Q\{S(Q)-1\} \sin(Qr)\, dr \quad \ldots\ldots\ldots\ldots \quad (4)$$

where n is the number of atoms per unit volume.

The radial distribution function is given by

$$Rh(r) = 4\pi r^2 g(r) \quad \ldots\ldots\ldots\ldots \quad (5)$$

The physical meaning of g(r) indicates the probability of finding an atom at the position r at the same time as we observe an atom at the origin. Thus the neutron diffraction measurements on amorphous materials allow us to measure the inter atomic distances of the constituent elements. The experimental S(Q) may be employed to obtain distribution function g(r) as well as radial distribution function Rh(r) of a system.

RESULTS AND DISCUSSIONS

The neutron diffraction data from amorphous quartz has been used to obtain the experimental structure factor, S(Q). The backgrounds at the initial part of the diffraction pattern have been subtracted. The experimental S(Q) as a function of Q has been shown in Figure 1. The experimental S(Q) has been used to calculate the g(r), the pair correlation function. The correlation function g(r) for amorphous quartz obtained by using experimental S(Q) is shown in Figure 2. In quartz there are two Si-O, two O-O and one Si-Si atom pairs. For crystal quartz the tetrahedron of oxygen atom about a silicon atom is almost regular, with Si-O = 1.61 A^o [13]. Besides its two silicon neighbors, each oxygen has six adjacent oxygens at distances ranging between 2.60 and 2.67 A^o. The amorphous quartz has some disorders in atomic arrangement apart from crystal quartz but the magnitude of disorder is not so high. The first two peaks in Fig. 2 indicate the Si-O and O-O atomic distances of the amorphous quartz sample. The distribution function g(r) obtained from present study has been compared with that of

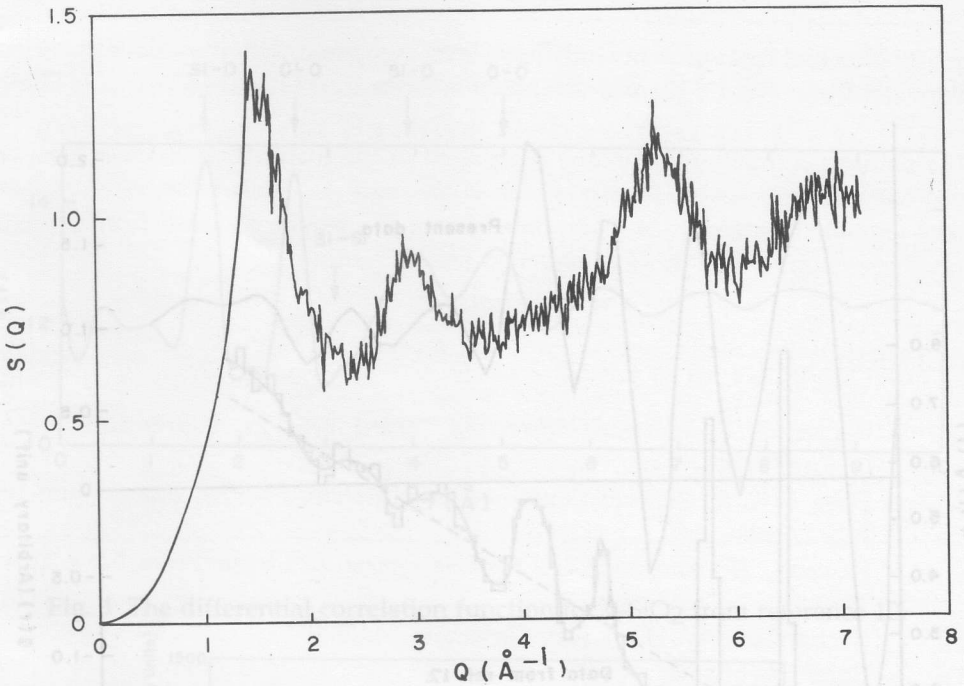

Fig. 1 The experimental S(Q) as a function of Q for amorphous quartz.

obtained from reference [12] and is also shown in Fig. 2. The agreement between them is good. M. Arai et al. [10] studied the dynamical pair correlation function of g-SiO_2 and compared it with differential correlation function g(r). The differential correlation function measured by them is shown in Fig. 3, from which the atomic distances have been calculated and compared with present results. Some measurements of neutron diffraction by amorphous silica have been made by Milligan et al. [11], who found six well-defined maxima, in contrast to a single broad peak and three ill-defined peaks revealed by X-rays. The resulting radial distribution curve has well-resolved peaks at 1.58, 2.56, 4.0 and 5.1 A^{o}. The radial distribution function obtained from the experimental data of the present study has also been shown in Fig. 4. The atomic distances of amorphous

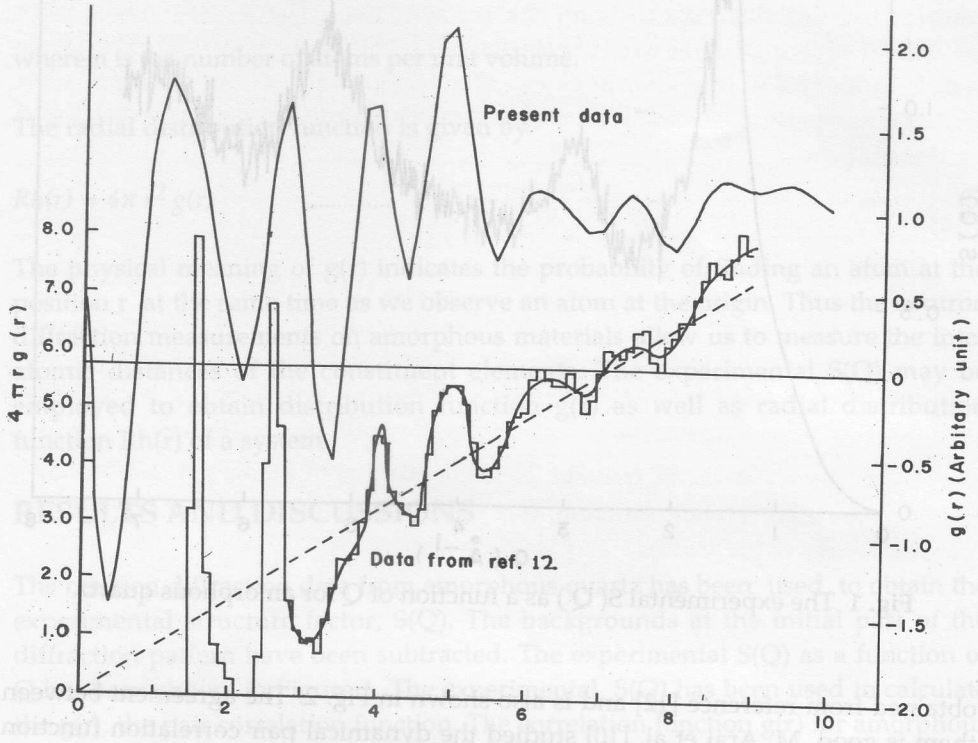

Fig. 2 The comparison of correlation function g(r) as a function of radial distance r for amorphous quartz.

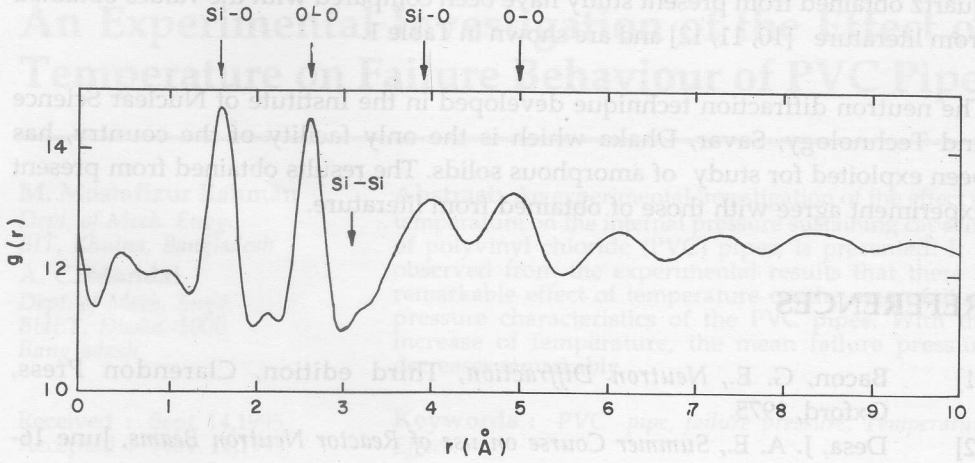

Fig. 3 The differential correlation function for g-SiO_2 from reference 10.

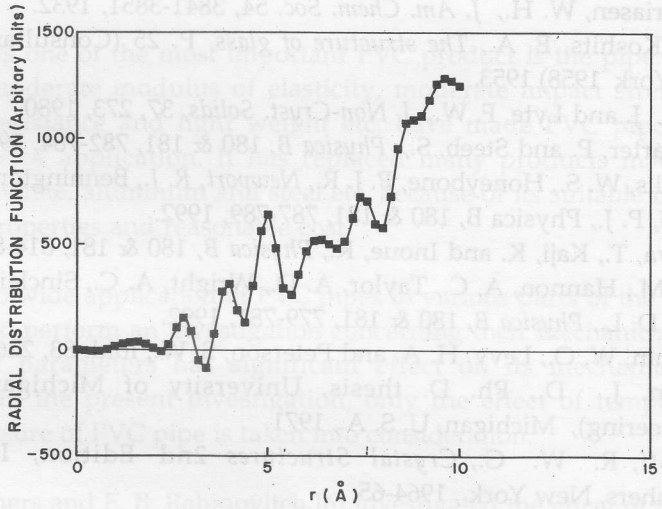

Fig. 4 The radial distribution function Rh(r) as a function r for amorphous quartz.

quartz obtained from present study have been compared with the values obtained from literature [10, 11, 12] and are shown in Table 1.

The neutron diffraction technique developed in the Institute of Nuclear Science and Technology, Savar, Dhaka which is the only facility of the country, has been exploited for study of amorphous solids. The results obtained from present experiment agree with those of obtained from literature.

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