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Medium range structure in a network glass established by a local probe

J. Wells [a], W.J. Bresser [b], P. Boolchand [b,*], J. Lucas [c]

[a] *Thomas More College, Crestview Hills, KY 41017, USA*
[b] *Department of Electrical and Computer Engineering and Computer Science, University of Cincinnati, Cincinnati, OH 45221-0030, USA*
[c] *Laboratoire des Verres et Céramiques, Université de Rennes, 35042 Rennes cedex, France*

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Abstract

Bulk Te_3Cl_2 and Te_3Br_2 glasses were prepared using Te tetrahalide and elemental Te as starting materials; they displayed glass transition temperatures of 357 and 348 K, respectively. In these stoichiometric chalcohalide glasses, a trimodal distribution of Te-cation sites is observed from ^{125}Te absorption and ^{129}I emission Mössbauer spectroscopies. The electric hyperfine structure reveals two of the sites (A, B) to be Te-intrachain sites characteristic of chains of c-Te_3Cl_2. The third site (A′), peculiar to the glasses, is identified as a chain terminator (or edge) site with a onefold-coordinated halogen atom. The average length of the chains in the glasses deduced from the observed Mössbauer site intensity ratios of the edge to interior (I'_A/I_A and I'_A/I_B) sites reveals a value of 1.5 nm in both glasses.

1. Introduction

Profound progress in decoding the nature of building blocks (short range order SRO) in network glasses has emerged from advances in experimental methods over the years. Elastic scattering methods (X-ray [1] and neutron pair-distribution functions [2]), vibrational spectroscopies (IR, Raman scattering [3], inelastic neutron scattering [4] and Lamb–Mössbauer factors [5]) and local methods (nuclear quadrupole resonance [6] (NQR), nuclear magnetic resonance (NMR) and magic angle spinning (MAS)-NMR [7] and Mössbauer effect [8]) each has provided complementary information on glass structure. In spite of these advances, the manner and extent to which local building blocks couple to form elements of medium range structure (MRS) in a glass continues to be speculative. Historically, the first sharp diffraction peak (FSDP) in the structure factor of network glasses has been widely regarded as signature of MRS, although its structural origin continues to be controversial [9–12]. Some [9–11] have ascribed the FSDP to a quasi-periodic arrangement of building blocks, as in a molecular cluster, extending over a characteristic length scale (cluster size). Elliott [12] has suggested that the commonality of this peak in a variety of glasses reflects a quasi-periodic distribution of free volume, i.e., inter-cluster correlations in the networks. This view has, however, not found support in recently reported first principles molecular dynamic simulations in glasses performed by Sarnthein et al. [12]. Raman scattering [13,14] in selective

* Corresponding author. Tel: +1-513 556 4758. Telefax: +1-513 556 7326. E-mail: pboolcha@ece.uc.edu.

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