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Amorphous and partly ordered structures in SiO_2 rich volcanic glasses. An ED study

VIKTÓRIA KOVÁCS KIS[1, 2,*], ISTVÁN DÓDONY[2] and JÁNOS L. LÁBÁR[1]

[1]Research Institute for Technical Physics and Materials Science, P.O.Box. 49, 1525 Budapest, Hungary
[2]Department of Mineralogy, Eötvös Loránd University, Pázmány Péter sétány 1/c, 1117 Budapest, Hungary
*Corresponding author, e-mail: kis@mfa.kfki.hu

Abstract: Glass structures of obsidian and pumice samples were measured using electron diffraction. Amorphous, partly ordered, and nanocrystalline regions were distinguished and analysed separately. The deconvoluted atomic distances obtained from experimental diffraction patterns through total pair-distribution functions are consistent with distances for ideal SiO_4 tetrahedra. Partly ordered structures in pumices are composed of plate-like fragments of tridymite/cristobalite layers, whereas obsidian contains quartz nanocrystals with abundant moganite-like planar faults. The validity of the structure-model of Goodman for silica glasses is discussed and an alternative interpretation is proposed for silicic volcanic glasses.

Key-words: glass structure, obsidian, pumice, electron diffraction.

Introduction

Volcanic glasses form by magma quenching. Silica-rich aluminosilicate magmas typically contain 72–75 wt% SiO_2, 13–15 wt% Al_2O_3, 1–4 wt% Na-, Ca-, K-oxides, OH groups and molecular H_2O. The physical, thermodynamical and rheological properties of aluminosilicate magmas are determined by their structures and compositions (*e.g.* Richet & Bottinga, 1995; Webb & Dingwell, 1995; Mysen, 2003). The nucleation and crystal growth properties of the submicron sized precipitates (mostly magnetite, pyroxenes, and amphiboles) called nanolites (Sharp *et al*., 1996) also depend on the structure of magma. Melt structures have been modelled in *ab initio* calculations (*e.g.* Sarnthein *et al*., 1995; Benoit *et al*., 2001; Pöhlmann *et al*., 2004). Direct measurements using diffraction techniques are hard to carry out. Studying volcanic glass structures allows us to deduce the structure of the parent aluminosilicate melt. Structural knowledge of magmas is essential in geology and volcanology (Dingwell, 1996).

Amorphous structures can be described by the nearest-neighbour interatomic distances, which can be inferred from diffraction data. These data can be transformed to a radial distribution function (RDF), which describes the variation of the atomic density, *i.e.*, the number of atoms in a spherical shell with thickness *dr*, at distance *r* from an origin chosen at any atomic position. The pair distribution function (PDF) shows the frequency distribution of individual bond distances between atoms, it is also effective in the quantitative description of highly disordered materials (see Gaskell, 1991).

Wright *et al*. (1984) made distinctions between silica (fulgurite) and silicate glasses (obsidian and tektite) based on neutron diffraction measurements. X-ray diffraction studies revealed that the structures of both obsidian and pumice are similar to the "stuffed tridymite" model proposed by Taylor & Brown (1979) (Zotov *et al*. 1989; Deganello *et al.,* 1998 and Zotov, 2003). Okuno *et al*. (1996) studied molten and untreated obsidian and also found that the basic structures of the molten and the untreated samples may be explained by the "stuffed tridymite" model. Beyond that Okuno and coworkers drew attention to the differences between RDFs of untreated and molten obsidians. In contrast to the untreated sample, the RDF function of the molten-and-quenched sample is featureless above the 5 Å region, which was interpreted by the authors as an outcome of a homogeneous, precipitate-free glass structure.

In fact, neutron and X-ray diffraction measurements on bulk samples do not provide a discrimination between the contributions of crystalline and amorphous components to the diffracted intensities.

In structural studies electron diffraction (ED) has some advantages of X-ray and neutron beam methods. The high spatial selectivity of ED allows excluding scattering contributions of precipitates to the pair distribution functions. The strong scattering power of electrons provides a short exposure time and good statistics in the signal. The light elements (the oxygen content is usually around 65 at%) are also well measurable.

We present a selected area electron diffraction (SAED) study on volcanic glasses. The ~0.2 μm spatial resolution of the SAED method allows to distinguish nanocrystalline,

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