Available online at www.sciencedirect.com

ScienceDirect

Acta Materialia 62 (2014) 250–257

Acta MATERIALIA

www.elsevier.com/locate/actamat

Constitutive modeling of the densification process in silica glass under hydrostatic compression

V. Keryvin [a,b,*], J.-X. Meng [a,b], S. Gicquel [b,1], J.-P. Guin [b], L. Charleux [b,c], J.-C. Sanglebœuf [b], P. Pilvin [a], T. Rouxel [b], G. Le Quilliec [b,2]

[a] Univ. Bretagne-Sud, EA 4250, LIMATB, F-56321 Lorient, France
[b] Univ. Rennes 1, ERL CNRS 6274, LARMAUR, F-35042 Rennes, France
[c] Univ. Savoie, EA 4144, SYMME, F-74940 Annecy-le-Vieux, France

Received 26 March 2013; received in revised form 24 July 2013; accepted 29 July 2013
Available online 28 October 2013

Abstract

The mechanical response of amorphous silica (or silica glass) under hydrostatic compression for very high pressures up to 25 GPa is modelled via an elastic–plastic constitutive equation (continuum mechanics framework). The material parameters appearing in the theory have been estimated from the ex situ experimental data of Rouxel et al. [Rouxel T, Ji H, Guin JP, Augereau F, Rufflé B. J Appl Phys 2010;107(9):094903]. The model is shown to capture the major features of the pressure–volume response changes from the in situ experimental work of Sato and Funamori [Sato T, Funamori N. Phys Rev Lett 2008;101:255502] and Wakabayashi et al. [Wakabayashi D, Funamori N, Sato T, Taniguchi T. Phys Rev B 2011;84(14):144103]. In particular, the saturation of densification, the increase in elasticity parameters (bulk, shear and Young's moduli) and Poisson's ratio are found to be key parameters of the model.

Keywords: Very high pressure; Silica glass; Modeling; Densification

1. Introduction

Because of their relatively low atomic packing density compared to their crystalline counterparts, glasses experience significant densification (permanent increase in density) under high hydrostatic pressures. In fact, the density of amorphous silica (a-SiO_2) can be increased by up to 20% and that of window glass by 6%, when a sufficiently high hydrostatic pressure is applied [1–6].

Permanent modifications in silica glass density are difficult to investigate via unconstrained macroscopic testing (such as the compression test) because of the material's brittleness. In contrast, hydrostatic compression on small volumes of material impede cracking drastically: permanent strains can be observed without any cracking features when possible spurious effects of additional shear are absent [7,8]. These tests usually give, after unloading (ex situ), information on the density changes. The combination of such tests with physical spectroscopy techniques (X-ray diffraction, Raman scattering, Brillouin scattering), e.g. in a diamond anvil cell, permits to follow in situ the changes in the structure of silica glass (short-to-medium range order) to be followed in situ. However, from a mechanical point of view, the in situ mechanical response of the test is partial as only the pressure information, and not the density information, is known during the test.[3]

Recent advances in experimental testing have made it possible to obtain the in situ mechanical response of the hydrostatic compression test (curve pressure–volume

* Corresponding author at: Univ. Bretagne-Sud, EA 4250, LIMATB, F-56321 Lorient, France.
E-mail address: vincent.keryvin@univ-ubs.fr (V. Keryvin).
[1] Present address: EDF – DPI, DCN/PÔLE Assemblage Combustible, 1, place Pleyel 93282 Saint-Denis Cedex, France.
[2] Present address: Univ. Tours, EA 2640, LMR, F-37000 Tours, France.

[3] It is, however, possible to extract this information assuming a purely elastic behaviour for pressures lower than the densification threshold [7].

1359-6454/$36.00
http://dx.doi.org/10.1016/j.actamat.2013.07.067

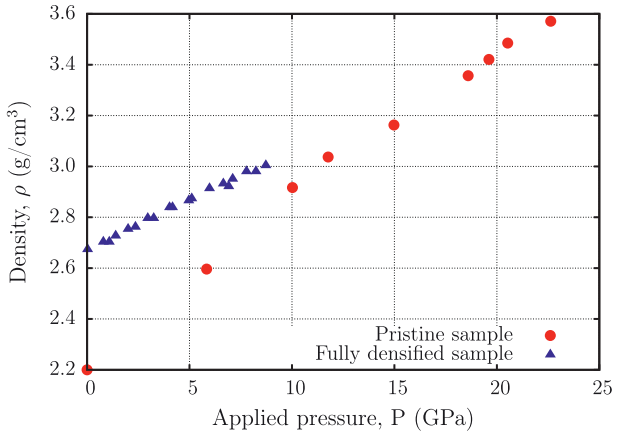

Fig. 1. Changes in density during in situ testing of silica glass under hydrostatic compression: pristine sample (circles) from Sato and Funamori [9] and fully densified sample (triangles) from Wakabayashi et al. [29].

changes). Sato and Funamori [9] conducted experiments up to 60 GPa with a diamond anvil cell at room temperature. The density of the silica glass sample was determined in situ from the intensities of transmitted X-rays measured for the sample and some reference materials (see Fig. 1). As well as this mechanical response, Sato and Funamori related their experiments to structure changes by using X-ray absorption and diffraction techniques [9,10]. They found that silica glass behaves as a single amorphous polymorph having a fourfold-coordinated structure below 10 GPa. Irreversible changes in the intermediate-range order begin at around 10 GPa (referred to as densification), up to 25 GPa. This corresponds to an irreversible and progressive transformation from a low-density amorphous phase to a high-density amorphous phase. This latter phase is characterized by an increase in the statistical distribution of four- and three-membered rings of SiO_4 tetrahedra with a narrowing of the inter-tetrahedral angle distribution [11,12].

From a more mechanistic point of view, the deformation mechanisms between 0 and 25 GPa may be depicted as follows [1–4,6,13]. Below a threshold pressure, the behaviour is purely elastic. Above a second threshold pressure, known as saturation pressure, the behaviour is once again purely elastic. In between these two pressures, densification occurs and develops by increasing the applied pressure (referred to as hardening[4]) and the elastic moduli increase with the densification level.

[4] Strain hardening is commonly ascribed to volume-conservative plasticity, where it is classically defined as the increase in flow stress upon plastic flow. Under such circumstances, the maximum strain is limited by the material strength. Otherwise, in the case of perfect plasticity, strains over 1 (superplasticity) could possibly be achieved because there is no geometrical constraint to shear processes. In contrast, densification is a geometrically constrained process, where pressure can be ideally increased to infinity without fracture, while strain is limited by the details of the atomic packing characteristics. It is obvious that densification becomes more and more difficult as the density increases. This has nothing to do with strain- or time-hardening processes observed in metal plasticity. Nevertheless, we will use the term hardening in this text.

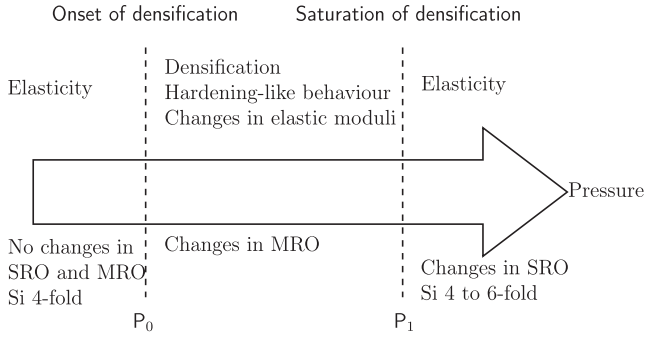

Fig. 2. Schematic of the deformation mechanisms in silica glass during hydrostatic compression (above the arrow) alongside the structural changes (below). The arrow stands for the increase in applied pressure. SRO and MRO refer to short range and medium range order, respectively. P_0 and P_1 are the onset and saturation pressures for densification, respectively.

Such a summary of structural changes, deformation mechanisms and constitutive models is found in Fig. 2.

Prior to this recent advance in experimental testing, the modelling of permanent deformation in glasses has been based on constrained mechanical tests that make it possible to develop stable permanent deformation fields without fracture or even cracking. This is the case, for instance, during hardness or scratch experiments. For temperatures well below the glass transition, according to the literature, the formation of the residual imprint is thought to result from the concomitant contribution of two deformation mechanisms: densification and shear flow [14–18,5,19]. Constitutive models were developed to clarify this issue on the hardness of glass [14,20–23]. They may involve only volume-conservative plasticity (further referred to as plasticity) – which is therefore unable to predict densification [14] – densification and plasticity [20–22], and even hardening [23]. The two latter models are based on the correct description of the instrumented indentation test response. The instrumented indentation test enriches the hardness test by giving access to the load vs. penetration curve. These new data are used to suggest more realistic constitutive equations in a straightforward way. The indentation test is heterogeneous by nature and, as a consequence, numerical simulations by the finite element method are generally used to link given material properties to the load vs. penetration curve and the residual imprint. Material parameters are then estimated using a identification procedure. Such models have been proposed for the last sixteen years, notably in the key works of Lambropoulos [22,24] and Kermouche et al. [23]. Both models assume that a combination of pressure and shear terms triggers permanent deformation (densification and plasticity). In all these models, attention has been paid mainly to the role of shear on the permanent deformation process. It appears, from the survey of these two constitutive models and the numerical simulations made with them, that different models allow one to fit load–displacements curves to the instrumented indentation [22,23]. It should be noted that some models