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Mechanical behaviour of fully densified silica glass under Vickers indentation

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ABSTRACT

When subjected to constrained deformation loadings, such as during an indentation test, silica glass experiences complex deformation mechanisms including densification and volume-conservative shear plasticity. The densification mechanism may increase the density up to more than 21%. The question of the mechanical behaviour of an already fully densified glass sample naturally arises. This issue is one of the key points to address when one tries to propose a constitutive model of pristine silica glass. Indeed, during an indentation test, which is a popular test for this task, beneath the indenter tip, some regions might be fully densified. What is their behaviour, after saturation in densification and during loading, is therefore an issue to address. Moreover, this is a crucial point for exploring the transition from plasticity to cracking, which is of paramount importance and a long-term objective for predicting the lifetime of glass products subjected to contact loadings such as impact or scratching.

In this paper, a quantitative identification of fully densified silica mechanical constitutive behaviour is made by using instrumented indentation testing and finite element analyses. The use of such an indirect method to assess the mechanical behaviour of this material comes from its brittle behaviour in unconstrained deformation modes usually employed in metals plasticity. It is shown here that fully densified silica behaves as a von Mises material (rate-independent shear plasticity without strain-hardening) like some crystalline metals. The yield strength and yield strains are, at the contrary, much higher than for alloys: respectively 6.5 GPa and 6.1%. This mechanical modelling, as well as the plastic parameters values found, are in excellent agreement with very recent experimental and numerical simulations on silica glass and silicate glasses.

The high value of the yield strain is found to explain unusual indentation features such as a unusual long range residual piling-up while sinking-in is predicted during loading, as well as low values of the ratio hardness-to-yield strength.

1. Introduction

Silica glass (SiO_2) is a highly brittle material that breaks in bending under few tens of MPa because of its extreme sensitivity to surface damage [1,2]. Yet, under constrained mechanical loadings such as hydrostatic tests or indentation, it can deform permanently without breaking or even cracking [3,4]. Under pure hydrostatic conditions, its density (respectively its volume) may increase permanently [5–7] by more than 20% (respectively decrease by more than 16%) [3,8]. This has been related to some changes in the intermediate range order, among which the decrease of inter-tetrahedra angles [9], and is referred to as pressure-induced densification (PID). It is now recognised that during indentation, densification occurs as well as volume conservative shear flow [10]. These main mechanisms at stake have been observed separately and very recently properly modelled: PID as recalled previously [11] and shear flow by uniaxial compression experiments [12,13]. For the former mechanism (PID), the authors proposed a description of the densification process with a threshold pressure for the onset of densification, an increase of densification upon pressure, and another pressure threshold for the saturation in densification. They also pointed out the necessity to use a Finite Deformation

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framework and to account for the elastic stiffening with densification (for instance the bulk modulus will double) [11]. As for the latter mechanism, the authors of Ref. [12,13], using compression tests on micropilars, showed that silica glass behaves in compression as a perfectly elasto-plastic material (without any strain hardening in shear) with a compressive yield strength around 6–7 GPa. These two mechanisms take place together during an indentation test [14,15] so that their kinetics must be related to a combination of driving forces (shear and pressure). Different authors have proposed such constitutive modellings that are successful in describing the mechanical response of the instrumented indentation test (the force-displacement curve) [16–20]. Some of them were also successful in observing densification fields beneath the residual imprint by Raman spectroscopy [18,21] or by a chemical dissolution technique [22].

However, a piece of this jigsaw puzzle is still missing: what does the behaviour of the material become when the densification is saturated to $\sim$ 21% ? In other words, what is the mechanical behaviour of fully densified silica glass? This issue is of paramount importance not only from a condensed-matter physics point of view but also since the ultimate target of modelling the mechanical response of glasses to surface damage is to describe the transition from plasticity to cracking and it is highly liable that these densified or fully densified zones play a major role in it.

This is the current objective of this paper. We will analyse instrumented indentation tests, in terms of force-displacement curves and residual imprints, to propose an adequate description of the mechanical behaviour of fully densified silica glass under Vickers indentation. The paper is organised as follows. We will first describe how silica glass samples are densified up to more than 21%, how indentation tests are performed and how results are analysed. The numerical procedures, using both two- and three-dimensional finite element analyses of the Vickers indentation test, will be then described. After presenting and analysing both experimental and numerical results, we will eventually challenge our findings in terms of mechanical modelling and material properties to very recent experimental and simulation literature results.

2. Experimental and numerical methods

2.1. Material and experimental procedures

Silica glass (Vitreosil™, Saint-Gobain, France) specimens (0.6 mm in diameter and 2 mm in length) were densified by means of an octahedral multi-anvil apparatus using a Walker cell. A typical run consisted in raising the load pressure of the main ram at a rate of 0.5 MPa/min of oil up to obtaining a pressure of 25 GPa on the sample. After reaching the target pressure, the specimens were maintained at high pressure for one hour and then slowly unloaded. Density was measured with a better than 0.001 g/cm^3 accuracy by means of a density gradient method using two partially miscible liquids (iodobenzene and methylene iodide). The density of the pristine samples was 2.2 g/cm^3. After unloading from the Walker cell, samples had a density increased by 21.6%. They are further referred to as fully densified (FD). Further details may be found elsewhere [3].

Instrumented indentation tests (IIT) were carried out with a micro-indenter testing device (Fischerscope H100 XYp, Fischer, Germany) at ambient conditions (23 °C and 55% relative humidity). It has a load resolution of 0.02 mN and a depth resolution of 2 nm. The calibration of the instrument was done by using ISO-14577 standard on a reference block (BK7™ borosilicate glass). The indenter tip is a Vickers diamond pyramid. IIT tests were carried out both on a pristine silica sample (further referred to as PS, studied for sake of comparison) and on FD. A typical '4-5-4' loading sequence was used: 4 s to reach the maximum force P_m, 5 s of holding time, and 4 s to unload the sample's surface. Tests were force-controlled and the P_m values were 100 mN. The mechanical response of the indentation test is the force P vs. the displacement δ (counted positively). Due to the high reproducibility of the indentation test on the glass surface, five indents per chosen maximum load were performed. Unloading curves were analysed to extract an indentation modulus, M, and an indentation hardness, H_{IT}, by Oliver and Pharr's method [23].

Atomic force microscopy made it possible to record the three-dimensional geometry of the residual indentation imprint after unloading. Images were captured by using the contact mode of an AFM (Bruker Nanoscope V, Dimension 3100, Santa Barbara, CA) equipped with silicon nitride tips (DNP) with a nominal tip radius of the order of 20 nm and apical angle of 40°. The AFM tip being much sharper than the Vickers indenter it ensures that the residual imprint shape is captured without AFM probe shape convolution effect. Finally, the piezo scanner was calibrated in X-Y-Z directions with a grid having a 10 μm pitch (X-Y directions) of 200 nm deep squared holes (Z direction).

Regarding image post treatment, the size of the scanned areas containing an indentation imprint was large enough so that a sufficient area, unaffected by the indentation process, surrounds the imprint and may be used as a reference surface for post treatment. This surface of reference was extracted from the raw image by using a disk-shaped mask centered on the imprint. It was assumed that the surface of the tested samples, far from the indentation imprint, was flat (i.e. no tilt and no offset). Thus a linear fit was applied to this data set, which was subsequently subtracted from the raw image. It is important to note that removing a tilt on the whole data set does not modify the three-dimensional shape of the residual imprint. Meyer's hardness, H, was also measured as the ratio P_m to the projected contact area, after unloading, on the residual imprints.

2.2. Numerical procedures

Finite element analyses (FEA) of the indentation process were performed using both two (2D) and three dimensional (3D) models constituted of a sample and an indenter. The 3D mesh is generated from a coarser 2D mesh. The 2D mesh is first described.

The sample's mesh is divided into a core zone, beneath the indenter tip, where the mesh is fine, and a shell zone where the mesh is coarse far from the contact. The core zone is itself divided into a square zone with a 32×32 (2D) or 16×16 (3D) quadrangular structured mesh contained into an outer unstructured zone made of quadrangular elements (32/16 again along the axis $z = 0$). The shell zone is decomposed into a transition zone where the element size is progressively increased and a outer zone, both with quadrangular elements. All elements are linear and use full integration.[1] The dimensions of the mesh are chosen in order to minimise the effect of the far-field boundary conditions. This is made by using a sufficient number of outer elements in the shell zone. The typical ratio of the maximum contact radius and the sample size is about 2×10^3. The 3D model relies on 45° swept 2D meshes since the Vickers pyramid has symmetries so that only 1/8th of it has to be meshed. The resulting elements are eight-node hexahedrons except along the axis where linear six-node prismatic elements are used. Detailed views of the meshes are shown in Fig. 1.

As for the indenter, the same procedure used for the sample

[1] No evidence of possible volumetric locking beneath the indenter tip has been observed.