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Fracture toughness anomalies: Viewpoint of topological constraint theory



Mathieu Bauchy^{a,*}, Bu Wang^a, Mengyi Wang^a, Yingtian Yu^a,
 Mohammad Javad Abdolhosseini Qomi^b, Morten M. Smedskjaer^c, Christophe Bichara^d,
 Franz-Josef Ulm^{e,f}, Roland Pellenq^{e,f,d}

^a Physics of Amorphous and Inorganic Solids Laboratory (PARISlab), Department of Civil and Environmental Engineering, University of California, Los Angeles, CA 90095, United States

^b Department of Civil and Environmental Engineering, Henry Samueli School of Engineering, University of California, Irvine, CA 92697, United States

^c Department of Chemistry and Bioscience, Aalborg University, 9220 Aalborg, Denmark

^d Centre Interdisciplinaire des Nanosciences de Marseille, CNRS and Aix-Marseille University, Campus de Luminy, Marseille, 13288 Cedex 09, France

^e Concrete Sustainability Hub, Department of Civil and Environmental Engineering, Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, MA 02139, United States

^f MIT-CNRS Joint Laboratory at Massachusetts Institute of Technology, 77 Massachusetts Avenue, Cambridge, MA 02139, United States

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ABSTRACT

The relationship between composition, structure, and resistance to fracture remains poorly understood. Here, based on molecular dynamics simulations, we report that sodium silicate glasses and calcium–silicate–hydrates feature an anomalous maximum in fracture toughness. In the framework of topological constraint theory, this anomaly is correlated to a flexible-to-rigid transition, driven by pressure or composition for sodium silicate and calcium–silicate–hydrates, respectively. This topological transition, observed for an isostatic network, is also shown to correspond to a ductile-to-brittle transition. At this state, the network is rigid but free of eigen-stress and features stress relaxation through crack blunting, resulting in optimal resistance to fracture. Our topological approach could therefore enable the computational design of tough inorganic solids, which has long been a “holy grail” within the non-metallic materials chemistry community.

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1. Introduction

Resistance to fracture, or fracture toughness, is of primary importance in materials science, with direct application to infrastructures and manufacturing. Indeed, tougher materials permit an increased resistance under extreme conditions, or the use of less materials while achieving comparable performances. To design tougher materials, one needs to understand the relationship between composition, structure, and toughness. This question has been identified as a “Grand Challenge” for non-metallic materials [1,2] since these solids (ceramics, cements, glasses etc.) are generally not able to undergo much plastic deformations, which inhibits the local dissipation of high stresses that cause the material to fracture. However, the fracture mechanism of these materials is

quite complex as, e.g., even archetypical brittle material like silicate glasses can exhibit some ductility at nanoscale [3–6]. In addition, as opposed to elastic moduli and hardness, which typically increase with connectivity and bond energy [7–9], toughness shows non-monotonic behaviors [10,7], which highlights the fact that fracture energy is not simply the energy required to break bonds and to create two new surfaces [10]. Tuning the fracture mechanism, e.g., through composition design, provides potential opportunities to improve the fracture resistance, but requires a unified model that can capture the mechanism transition between ductile and brittle behaviors and predict its compositional dependence for a broad range of materials.

To this end, topological constraint, or rigidity theory [11–14] appears to be an efficient tool. The theory reduces the complex atomic structure of materials into a simple mechanical truss, in which the atoms are connected to each other through topological constraints, that is, the chemical interactions that constrain the values of bond lengths and bond angles. Following this framework

* Corresponding author.

E-mail address: bauchy@ucla.edu (M. Bauchy).

and inspired by Maxwell's study on the stability of mechanical trusses [15], rigidity theory has been extensively used to understand the compositional dependence of the properties of network glasses. As such, an atomic network is classified as *flexible*, having internal degrees of freedom that allow for local deformations [16], *stressed–rigid*, being locked by its high connectivity and featuring eigen-stress [17,18], or *isostatic*, the optimal intermediate state, rigid but free of eigen-stress. The isostatic state is achieved when the number of constraints per atom n_c , comprising radial bond-stretching (BS) and angular bond-bending (BB), equals three, the number of degrees of freedom per atom. Compositions offering an isostatic behavior have been found to exist inside a window [19], located between the flexible ($n_c < 3$) and the stressed–rigid ($n_c > 3$) domains, and show some anomalous properties such as space-filling tendency [20,21], weak aging phenomena [22], and anomalous dynamical and structural signatures [23–26]. Rigidity theory has been shown to be a powerful tool for predicting the mechanical properties of glass [8,9,27] and has been used to design the Corning® Gorilla® Glass 3 [28–30], a scratch- and damage-resistant glass used on more than one billion smartphones and tablets screens.

Here, we investigate two families of materials that suffer from low fracture toughness, densified sodium silicate glasses (NS) and calcium–silicate–hydrates (CSH). NS is a base material of various multi-components silicate glasses found in industry (e.g., window glasses) as well as in geoscience (e.g., magmas [31]). On the other hand, CSH is the binding phase of concrete, a ubiquitous material in our built environment, and is responsible for its mechanical properties [32]. Based on molecular dynamics (MD) simulations, we report anomalous fracture toughness behaviors for both materials. Interestingly, in both cases, a maximal resistance to fracture is observed for isostatic networks, analog to the reported data of germanium-selenium (GS) chalcogenide glass. We find that, although these materials substantially differentiate in the nature of their structure and chemistry, rigidity theory ubiquitously predicts the mechanistic transition in the fracture behavior as a function of their atomic topology. We therefore expect it to provide a versatile tool for efficient composition design of tougher materials.

2. Methods

The topology and resistance to fracture of permanently densified NS glasses and CSH of different compositions are investigated through MD simulations. The atomic configurations of these systems are prepared and validated relying on the following procedure.

The permanently densified $(\text{Na}_2\text{O})_{30}(\text{SiO}_2)_{70}$ samples are constructed via quenching liquid melts consisting of 3000 atoms at constant selected pressures. The simulations use a classical potential set developed by Teter [33] that has been extensively studied and shown to provide realistic results for the structural, dynamical, and mechanical properties of NS glasses and super-cooled liquids [31,33–36]. Following the procedure described in Ref. [37], the quenching procedure is performed by equilibrating the melts at 3000 K for 1 ns to lose the memory of the initial configurations, and linearly cooling the melts from 3000 to 300 K at a cooling rate of 1 K/ps and under constant pressure to form permanently densified glasses. When the glass is quenched under a series of pressure from 0 to 22 GPa, the rigidity of the structure increases as the densification induces an increase in the coordination number of the network-former atoms, that is, one observes the formation of five- and six-fold coordinated Si and three-fold “tricluster” O atoms [38]. Prior to fracture simulations, the obtained NS glasses are relaxed to zero pressure, but these highly coordinated species are retained and the glasses remain

permanently densified.

On the other hand, the CSH samples $(\text{CaO})_x(\text{SiO}_2)(\text{H}_2\text{O})_y$ are generated by introducing defects in an 11 Å tobermorite [39] configuration, following a combinatorial procedure [40,41]. 11 Å tobermorite consists of pseudo-octahedral calcium oxide sheets, which are surrounded on both sides by silicate tetrahedral chains, made of BO atoms and Q^2 silicon atoms [42,43]. These negatively charged calcium–silicate sheets are separated from each other by an interlayer spacing, which contains water molecules and charge-balancing extra calcium cations [44]. In the present models, starting from 11 Å tobermorite, the Ca/Si molar ratio is increased from 1.0 to 1.9 through randomly removing SiO_2 groups, to reach the range of compositions observed experimentally for CSH [41]. The defects in the silicate chains provide possible sites for the adsorption of extra water molecules, which is performed through Grand Canonical Monte Carlo simulations. The computed amount of adsorbed water is in excellent agreement with experimental data, which reflects the relevance of the present approach in describing CSH [41]. The ReaxFF potential [45], a reactive potential, is finally used to account for the reaction of the interlayer water with the defective calcium–silicate sheets, thereby creating hydroxyl groups [46]. The details of the simulations of the CSH samples, as well as their hardness computation, can be found in [Supplementary Note 1 and 3](#) and Ref. [41].

After realistic atomistic samples are obtained, topological constraint analyses and simulations of fracture are performed, the details of which are reported in the results section.

3. Results

To investigate the atomic topology and resistance to fracture, we reply on realistic MD simulations of permanently densified NS glasses and CSH of different compositions (see the Methods section). These systems are interesting as, in addition to their industrial relevance, their atomic networks feature varying degree of connectivity with respect to the pressure during quenching and composition, respectively.

As discussed in the Introduction, the mechanical constraints experienced by the atoms inside a molecular network include radial BS and angular BB constraints. Counting the number of constraints has been well established in fully connected glasses featuring covalent bonds like $\text{Ge}_x\text{Se}_{1-x}$ (GS). In such glasses, each atom experiences $r/2$ BS constraints (note that each BS constraint is shared by two atoms) and $2r - 3$ BB constraints, where r is the coordination number [14]. On average, the number of atomic constraints per atom is then given by $n_c = \langle r \rangle / 2 + (2\langle r \rangle - 3)$, where $\langle r \rangle$ is the average coordination number. In GS, Ge and Se atoms feature a coordination number of 4 and 2, respectively, so that the rigidity of the network (n_c) can be evaluated from the mere knowledge of the composition. However, in more complex materials, the enumeration of the constraints requires care, as their coordination numbers are not necessarily known. Moreover, it has been shown that the constraints can be intact or broken depending on the temperature [47,48]. To tackle this issue, we refine a method originally developed in Ref. [49], and widely applied to chalcogenide and oxide glasses since then [23–26]. The enumeration is based on the analysis of atomic trajectories obtained through molecular dynamics simulations, from which the numbers of BS and BB constraints are computed by computing the standard deviations σ of the bond lengths and angular distributions, respectively. The basic idea is that intact constraints induce low σ , whereas broken ones result in large σ . The constraints enumeration method used in NS and CSH can be found in Refs. [37] and [50], respectively, and clearly distinguishes intact from broken constraints. As shown in Fig. 1, both NS and CSH show a rigidity transition. Indeed, NS becomes

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