Glass structure, melt structure, and dynamics: Some concepts for petrology

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ABSTRACT

The thermodynamic and transport properties of the aluminosilicate melts at the heart of most magmatic processes vary in complex ways with composition, temperature, and pressure. Insights into these properties can come from information on the structure of the melts themselves, and more commonly from their glassy, quenched equivalents. Although most such connections remain qualitative or semi-quantitative, they are fundamentally important in interpretation of observations on igneous systems in nature and the laboratory, and in the formulation of physically accurate models. This review presents some of the important concepts of aluminosilicate glass and melt structure and dynamics that are most relevant to furthering our understanding of the igneous processes so central to how our planet has formed and evolved. The relationships among glasses, melts, and crystals are introduced. The structural underpinnings of temperature and pressure effects on melt free energies, densities, and viscosities, constraints on the extent of order/disorder among cations and anions in melts, why silica activity varies so strongly with composition, and how liquid-liquid phase separation can be understood, are discussed. Some simple, but useful, general views are presented on melt disorder and the shapes of liquidus surfaces (key to magmatic phase equilibria), as are links between atomic-scale dynamics and viscous flow and diffusion.

Keywords: Silicate melt, glass structure, phase equilibria, viscosity, igneous process, Invited Centennial article, Review article

INTRODUCTION

After decades of research using many experimental and theoretical approaches, much is now known about the structure of aluminosilicate glasses. These materials are of interest not only to igneous processes in nature, but in advanced technologies as well. Less well-defined are the high-temperature (and often high pressure) liquids themselves, but work on glasses has provided a basic framework, and growing numbers of in situ, high T, high P, and even high P-T experiments are becoming feasible. However, as may be inevitable in a developing field, the specialized language and concepts of the discussion, and the focus on ever-finer quantitative details, can limit accessibility by non-experts. This can, in turn, limit motivating inputs from those studying petrological processes into future directions of investigations on the atomic scale. Conversely, phenomenological models that are generated to make pragmatic predictions of melt properties, such as compositional fits of phase equilibria or physical property data, may have limited physical accuracy if they are not well informed by structural concepts. And, a more general understanding, by a wider scientific community, of the fundamental roots of well-known petrological phenomena may lead to new insights into how igneous processes occur in nature as well as the laboratory or industrial glass production facility. Increasingly accurate, and more automated, tools have been developed for calculating the behavior of melts, which often now appear as “black box” software packages. The very success of such modeling efforts can actually insulate users from underlying connections between structure, atomic-scale process, and melt properties, obscuring fundamental and intriguing questions. For example, why do the heats and entropies of fusion for minerals, buried in a model algorithm, vary so greatly, and why does this matter so much for geological systems?

It is thus the purpose of this review to present some of the basic concepts of melt structure and properties in a brief, non-technical form that may increase their accessibility to students and researchers of real-world igneous processes, and enhance the two-way cross-fertilization that can enhance progress across the entire spectrum. Given the breadth of this field, and the limitations of a relatively short article, not much detail can be provided. Recent introductions to the field (Calas et al. 2006; Dingwell 2006; Galoisy 2006; Henderson et al. 2006), as well as, more extensive reviews, provide not only greater conceptual depth but the background literature from experiment and theory that lies beneath the generalizations given here (Greaves and Sen 2007; Lee et al. 2004; Mysen 1990; Mysen and Richet 2005; Richet and Bossinga 1986; Richet and Neuville 1992; Stebbins et al. 1995a, 2013; Stebbins and Xue 2014). Several areas of particular interest in petrology, where some real progress has been made in connecting melt structure with macroscopic properties, are also neglected here for length reasons, notably the role of volatile components such as H2O and crystal-melt partitioning. Insights from quantum chemical calculations and computer simulations, which have long been important in thinking about melt structure...