



Impact of iron chelators on short-term dissolution of basaltic glass

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Abstract

Although microorganisms seem to play an important role in the alteration processes of basaltic glasses in solution, the elementary mechanisms involved remain unclear in particular with regard to the role of organic ligands excreted by the cells. Two glasses, one with Fe and one without Fe were synthesized to model basaltic glass compositions. Fe in the glass was mostly Fe(III) for enhancing interaction with siderophores, yet with small but significant amounts of Fe(II) (between 10% and 30% of iron). The prepared samples were submitted to abiotic alteration experiments in buffered (pH 6.4) diluted solutions of metal-specific ligands, namely oxalic acid (OA, 10 mM), desferrioxamine (DFA, 1 mM) or 2,2'-bipyridyl (BPI, 1 mM). Element release from the glass into the solution after short term alteration (maximum 1 week) was measured by ICP-OES, and normalized mass losses and relative release ratios (with respect to Si) were evaluated for each element in each experimental condition. The presence of organic ligands had a significant effect on the dissolution of both glasses. Trivalent metals chelators (OA, DFA) impacted on the release of Fe³⁺ and Al³⁺, and thus on the global dissolution of both glasses, enhancing all release rates and dissolution stoichiometry (release rates were increased up to 7 times for Al or Fe). As expected, the mostly divalent metal chelator BPI interacted preferentially with Ca²⁺, Mg²⁺ and Fe²⁺. This study thus allows to highlight the central roles of iron and aluminium in interaction with some organic ligands in the alteration processes of basaltic glasses. It thus provides a step toward understanding the biological contribution of this fundamental geological process.

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1. INTRODUCTION

Alteration mechanisms of glasses constitute a major area of interest in many research fields. In an environmental

context, studies about nuclear glass weathering have been largely reported in the literature. Short-term alteration experiments have been designed to determine initial dissolution rates (Lucktscheiter and Nesovic, 2004; Fournier et al., 2014) while residual dissolution rates have been calculated for very long-term alteration experiments (Curti et al., 2006; Libourel et al., 2011; Gin et al., 2012, 2013, 2014). These short and long-term dissolution kinetics were investigated with respect to parameters such as temperature and pH (Pierce et al., 2008) and glass elemental composition

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(Gin et al., 2012), and they have been coupled to studies of the alteration gel (Rebiscoul et al., 2005; Jollivet et al., 2008; Gin et al., 2015; Hellmann et al., 2015) or of secondary phases (Pelegri et al., 2010). These experiments were performed in pure water, water enriched in elements from the glass (Luckscheiter and Nesovic, 2004; Thien et al., 2012) and in salt solutions (Godon et al., 1988; Abdelouas et al., 1993). The stability and dissolution behavior of bioglass, designed to form chemical bonds with living tissues (Souza et al., 2013), is of interest to the medical field. Glass alteration is also a crucial issue in the field of cultural heritage, especially with respect to the restoration and conservation of stained glass windows (see, for example Silvestri et al., 2005; Warkinson et al., 2005; Cagno et al., 2011; Gentaz, 2011; Schalm et al., 2010; Sterpenich, 2011; Lombardo et al., 2013). Finally, in a geological context, igneous rock alteration processes and especially basaltic glass-fluid interactions have been examined, as they provide knowledge about the major biogeochemical cycles of the Earth (Fliegel et al., 2012; Knowles et al., 2013). For example, glass weathering plays a central role in geological sequestration of CO₂ (Knauss et al., 2005) and contributes to the cycling of elements such as calcium, magnesium, silicon, iron and also oxygen in soils, rivers, lakes and oceans (Gislason et al., 2009). Basaltic glasses are also considered to be analogues of some nuclear glasses (Crovisier et al., 2003; Parruzot et al., 2015) and by extension, of archaeological glasses (Verney-Carron et al., 2008; Michelin et al., 2013).

The dissolution mechanisms of silicate glasses in solution have been extensively documented in the literature (Techer et al., 2000; Oelkers, 2001; Stroncik and Schmincke, 2002; Crovisier et al., 2003; Verney-Carron et al., 2008; Fournier et al., 2014). However, few studies have investigated this alteration under the action of microorganisms (Drewello and Weissmann, 1997; Gorbushina and Palinska, 1999; Gallien et al., 2001; Aouad et al., 2006; Stockmann et al., 2012; Shen et al., 2014) despite their well known interaction with mineral surfaces in general (Hutchens, 2009). Microorganisms have been shown to either enhance or inhibit dissolution of most minerals by a variety of mechanisms, especially through a direct impact of bacteria attached to the glass surface (Hutchens, 2009) and also by considering the possible effect of microbial metabolites excreted by the cells (Ullman et al., 1996). An example of this is siderophores, which are excreted in response to iron deficiency by different biological sources such as bacteria or fungi, in soils and in oceans. They have been shown to target and acquire specifically iron from iron-containing minerals and to mediate metal transport and re-absorption into the cell (Kalinowski et al., 2000b; Kraemer, 2004; Wolff Boenisch and Traina, 2007). As a consequence, they have been described as catalytic agents of the dissolution of those minerals. A considerable number of studies have described this ligand-controlled dissolution as a surface controlled process to which an empirical rate law can be applied:

$$R_t = R_H + R_L \quad (1)$$

where R_t is the overall dissolution rate, R_H the proton-promoted rate and R_L a ligand-promoted term (Furrer and

Stumm, 1986; Welch and Ullmann, 1993; Drever and Stillings, 1997; Stillings et al., 1998; Rosenberg and Maurice, 2003; Cama and Ganor, 2006).

Iron is a micronutrient that is essential for a range of crucial enzymatic processes in most organisms. In most environments iron deficiency is triggered by low iron bioavailability. To overcome this limitation, microorganisms and especially bacteria are known to sequester iron using organic molecules (Kraemer, 2004). Most siderophores are Fe³⁺-ligands, but they are most of the time able to bind ions other than Fe³⁺ and notably divalent cations (Hernlem et al., 1996; Braud et al., 2009; Brandel et al., 2012). Although considerable research has been carried out to examine the processes and products of abiotic basaltic glass weathering, little work has been done to quantitatively understand their weathering process in the presence of bacteria and organic bioproducts (Staudigel, 1995).

In this work, the impact of siderophores on the alteration of two synthetic basaltic glasses was investigated as a first step towards the understanding of natural systems. Three iron chelators, namely oxalic acid (OA), desferrioxamine (DFA) and 2,2'-bipyridyl (BPI) were chosen on the basis of their respective affinity with iron in its Fe(II) and/or Fe(III) oxidation states. The effect of OA on mineral dissolution has been reported in the literature (Furrer and Stumm, 1986; Zinder et al., 1986; Welch and Ullman, 1992; Stillings et al., 1995; Drever and Stillings, 1997; Oelkers and Gislason, 2001; Cheah et al., 2003; Wang et al., 2005; Olsen and Rimstidt, 2008; Martinez-Luevanos et al., 2011). OA can be complexed with both ferric and ferrous ions but its affinity with trivalent metal cations is stronger. DFA is a hexadentate siderophore produced by the soil bacterial strain *Streptomyces pilosus*, in which three hydroxamate groups, each acting as a bidentate ligand, contribute to the specific ligation of Fe³⁺ and therefore to the formation of very stable 1:1 complexes with aqueous Fe(III) (Kraemer et al., 1999; Liermann et al., 2000; Elandaloussi, 2003). By contrast, BPI is a bidentate chelator that predominantly binds aqueous Fe(II). Three molecules of BPI are necessary to coordinate with one Fe atom. While the impact of DFA on dissolution of mineral phases has been widely investigated in studies which illustrate the synergistic effect that the molecule has on dissolution (Watteau and Berthelin, 1994; Kraemer et al., 1999; Liermann et al., 2000; Cocozza et al., 2001; Cheah et al., 2003; Wolff-Boenisch, 2007), the interactions between 2,2'-bipyridyl and minerals have been the goal of only a few studies mainly focusing on the adsorption process of BPI on solids surfaces (Coluccia et al., 1978; Ferreira et al., 1983).

The use of Fe-specific chelators implies that we are focussing on the solvation of Fe and its possible impact on the global dissolution of the glass. In this regard, two different glasses (with and without Fe) were prepared using a simplified basaltic glass composition. Experiments were performed to evaluate the significance of the ligand-promoted dissolution and to determine the effect of three organic ligands, namely OA, DFA and BPI, on basaltic glass dissolution mechanisms at pH 6.4 and 25 °C. Experimental conditions were chosen to ensure a negligible

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