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A metasomatic mechanism for the formation of Earth's earliest evolved crust

Don R. Baker ∗, Kassandra Sofonio

Earth and Planetary Sciences, McGill University, 3450 rue University, Montréal, QC H3A 0E8, Canada

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Following giant impacts the early Hadean Earth was shrouded in a steam atmosphere for durations on the order of 1 Ma. In order to investigate the potential of this atmosphere to fractionate major elements between various silicate reservoirs and influence a planet's geochemical evolution, we performed experiments simulating the interaction of a post-giant-impact steam atmosphere with a bulk silicate Earth (BSE) composition. Our experiments indicate that the composition of the solute in a water-rich atmosphere at 10 MPa and ∼727 ◦C is remarkably similar to that of Earth's modern continental crust and would constitute up to 10% of the solution mass. This solute composition is similar to solute compositions previously measured at higher pressures, but distinct from those of near-solidus peridotite melts. Mass balance calculations based upon the hypothesis that Earth's initial water concentration was similar to that in CI carbonaceous chondrites, and that degassing and metasomatism produced the BSE, indicate that metasomatism could produce from 10 to 300% of the mass of the modern crust. If instead the amount of metasomatism is estimated by the difference between the water concentration in the BSE and in the depleted upper mantle, then a mass of up to approximately 4% of the current crust could be produced by metasomatism. Using results of earlier research we find that the solute is expected to have a smaller Sm/Nd ratio than the residual BSE, and if the solute was formed early in Earth's history its Nd isotopic signatures would be highly enriched. Although we cannot be certain that the metasomatic process created a significant fraction of Earth's crust in the early Hadean, our research indicates that it has the potential to form crustal nuclei and possibly was responsible for the production of incompatibleelement enriched reservoirs in the early Earth, as seen in the isotopic signatures of Archean rocks.

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1. Introduction

The first hundred million years of Earth history was a tumultuous time characterized by intense bombardment and differentiation of the planet. The proto-Earth was bombarded with planetoids and planetesimals, including a Mars-sized planetoid whose collison was responsible for the formation of Earth's moon and melting of the mantle to create an ultramafic magma ocean [\(Hartmann](#page--1-0) and Davis, [1975; Cameron](#page--1-0) and Ward, 1976). During early Hadean time, metal and silicate differentiation created Earth's core and multiple silicate reservoirs of differing composition (e.g., [Bennett](#page--1-0) et al., [2007; Moynier](#page--1-0) et al., 2010; Carlson et al., 2014). Rapidly, this environment cooled into a world where zircons could crystallize in granitic magmas at conditions similar to those of modern granites only 100 Ma later [\(Harrison,](#page--1-0) 2009). Understanding this earliest

Corresponding author.

E-mail addresses: don.baker@mcgill.ca (D.R. Baker), kassandra.sofonio@mail.mcgill.ca (K. Sofonio).

epoch of Earth's history will advance our knowledge of the evolution of terrestrial planets and serve as a guide in the search for exoplanets that may harbor life.

Early silicate differentiation processes played fundamental roles in determining the composition of the depleted mantle and incompatible-element-enriched reservoir or reservoirs, as revealed by isotopic measurements of younger rocks and crystals (e.g., Harper and [Jacobsen,](#page--1-0) 1992; [Bennett](#page--1-0) et al., 1993, 2007; [Boyet](#page--1-0) et al. [2003;](#page--1-0) Caro et al., [2006; Harrison](#page--1-0) et al., 2008; O'Neil et al., 2008; Moynier et al., [2010; Rizo](#page--1-0) et al., 2012). The processes responsible for silicate differentiation remain under discussion because of the lack of any rock or crystal samples dating from the earliest stages of Earth's history, but fractional crystallization of a magma ocean, or of near-surface melts created by large impacts, are cited as possible mechanisms (e.g., Boyet et al., [2003; Darling](#page--1-0) et al., 2009; Rizo et al., [2012; Carlson](#page--1-0) et al., 2014). Although these mechanisms appear inevitable outcomes of large impacts on the early Earth, another effect of impacts on a terrestrial planet that plays a role in silicate differentiation has not been sufficiently considered. This process is the metasomatic interaction of high-temperature

Fig. 1. Schematic (and most definitely not to scale) of aerial metasomatic conditions on a terrestrial planet approximately 1 Ma after a giant impact following the model of Lupu et [al. \(2014\).](#page--1-0) Shown in the schematic is the convecting and degassing magma ocean beneath a thin veneer of mafic-to-ultramafic crust with a 10 MPa steam atmosphere above it. At the time considered in this schematic only a small-portion of the near-surface Earth was undergoing convection. At this time the planet is still bombarded by large impactors with enough energy to vaporize some rock material (left hand side) and smaller, less-energetic, ones (right hand side). The surface temperature is approximately 1000 K and the lower atmosphere is reacting with silicates on the surface and in the air. At higher levels in the atmosphere condensation of silicate material and water create a silicate rain that may evaporate in the lower, hotter portions of the atmosphere. The colors (online only) of the mantle and the near-surface atmosphere in this figure correspond to those of black bodies at temperatures of ∼1500 and 1000 K, respectively.

carbon–oxygen–hydrogen, C–O–H, fluids with silicates on Earth's surface and in post-impact atmospheres, a process we refer to as aerial metasomatism to distinguish it from classical, metamorphic metasomatism in Earth's crust and mantle (Fig. 1).

Aerial metasomatism occurs late in the sequence of events associated with large impacts. The impact itself begins the sequence and its subsequent effects depend upon the size of the impactor. A Mars-sized impactor $(6.8 \times 10^{23} \text{ kg})$ can vaporize all the silicate Earth and heat it to 3690 K whereas an impactor of the size of the asteroid 2 Pallas $(1.4 \times 10^{20} \text{ kg})$ only boils the oceans and creates maximum temperatures of approximately 2000 K [\(Fegley](#page--1-0) and [Schaefer,](#page--1-0) 2014). A giant impact, the size of the Moon-forming one, deposits enough energy into the Earth to create a silicate atmosphere that exists for 10^3 to 10^4 y (Fegley and [Schaefer,](#page--1-0) 2014 and references therein). After this time, most of the silicate materials condense out of the atmosphere and rain onto the planet, possibly producing deposits similar to the spherule beds made by smaller, early Archean, impacts (Lowe and [Byerly,](#page--1-0) 1986). The silicate atmosphere is replaced by one dominated by water for oxidized conditions, or mixtures of $CO₂$, H₂, and CO for reduced conditions (cf. [Schaefer](#page--1-0) and Fegley, 2010). As the planetary surface cools below the peridotite liquidus in \sim 2 × 10⁵ y after the impact [\(Lupu](#page--1-0) et al., [2014\)](#page--1-0) a "tenuous solid rind" [\(Sleep](#page--1-0) et al., 2001) forms. Cooling of surface temperatures to near 0 ◦C are calculated to occur between ~1.6 × 10⁶ y (Lupu et al., [2014\)](#page--1-0) and 2.5 × 10⁶ y [\(Sleep](#page--1-0) et al., [2001\)](#page--1-0) after the impact, although the cooling models do not precisely constrain the time during which the surface cools from ∼1200 ◦C to near 0 ◦C (e.g., [Sleep](#page--1-0) et al., 2015). During this cooling the atmospheric pressure on the planet has been modeled to range from 10 to 100 MPa (e.g., Sleep et al., [2001, 2015;](#page--1-0) [Lupu](#page--1-0) et al., [2014\)](#page--1-0). As the planet continues to be pummeled by planetoids and planetesimals after a giant impact, the more-common, smaller, ocean-vaporizing impacts could each create steam atmospheres lasting for thousands of years [\(Sleep](#page--1-0) et al., 1989) that would also be metasomatic agents. The atmospheres would dissolve silicate material at high temperatures in the lower atmosphere that might condense at higher, cooler levels as a silicate rain. Aerial metasomatism occurs from the condensation time of a silicate atmosphere to the condensation of a steam atmosphere (as it is traditionally named, see Fegley and [Schaefer,](#page--1-0) 2014) through interactions of the high temperature fluids in the atmosphere and silicate material suspended in the atmosphere or on the planetary surface (Fig. 1). As the atmosphere cools to temperatures low enough to allow water to condense, any silicate material formerly dissolved in the steam atmosphere will rain onto the surface.

Traditional metasomatism could operate simultaneously with aerial metasomatism. The vigorously convecting magma ocean beneath the rind will exsolve most of its volatile content as it ascends due to the positive correlation between water solubility in silicate melts and pressure [\(Sleep](#page--1-0) et al., 2015). As these volatiles move through the rind on the planet's surface and into the atmosphere they also act as metasomatic agents transporting material from mantle depths to the near-surface environment and atmosphere. However, even in this case equilibrium is expected to occur at near-surface conditions and the effects will be similar to aerial metasomatism.

The consensus that a high-temperature, C–O–H atmosphere surrounds a post-giant-impact Earth (e.g., Sleep et al., [2001,](#page--1-0) 2015; Schaefer and Fegley, 2010; Abe, [2011; Fegley](#page--1-0) and Schaefer, 2014; Lupu et al., [2014\)](#page--1-0) raises the question of its role in the silicate differentiation of a planet. Thermodynamic models of aerial metasomatism of a magma ocean at temperatures of 2000 to 3000 K were reported by Fegley et [al. \(2016\).](#page--1-0) We, instead, investigated metasomatism occurring at lower temperatures that exist for significantly longer time periods than that during which a magma ocean interacts with the atmosphere (Fegley and [Schaefer,](#page--1-0) 2014; Lupu et al., [2014;](#page--1-0) [Sleep](#page--1-0) et al., 2015). We experimentally investigated the role of metasomatism in silicate differentiation by studying the major element dissolution of a bulk silicate Earth, BSE, composition into hydrous fluids at conditions similar to those used in the modeling of Lupu et [al. \(2014\).](#page--1-0)

2. Experimental and analytical methods

Lupu et [al. \(2014\)](#page--1-0) modeled the thermal evolution and atmospheric composition of a planet following a giant impact; one of their scenarios used a surface pressure of 10 MPa and a surface composition of the BSE for the planet. A pressure of 10 MPa is similar to that produced by vaporizing the current oceans on Earth (Abe et al., [2000\)](#page--1-0). Performing experiments at 10 MPa and the upper end of the temperature range (1500 \degree C and higher) of the postimpact steam atmosphere is challenging because of the need to use large capsules while maintaining isothermal conditions. Because of the available experimental apparatus we chose a temperature at the middle of the range (and near the safety limit of our experimental apparatus), ∼727 ◦C, or 1000 K, to experimentally simulate metasomatism at a pressure of 10 MPa.

A mixture of bulk silicate earth composition, based upon that of Palme and [O'Neill \(2014\),](#page--1-0) was created from reagent-grade oxides and carbonates mixed for 17 h in a rotary mixer and then decarbonated overnight at 990 ◦C. The starting material was melted twice at 1550 °C with intermediate grinding. After the second melting the mixture was ground to a powder and stored in a 110 \degree C oven. This starting material contains crystals of olivine, orthopyroxene, clinopyroxene and plagioclase plus a small amount of glass; its composition is given in [Table 1.](#page--1-0)

Experiments were performed in gold palladium ($Au_{75}Pd_{25}$) double capsules. Each assembly consists of an outer capsule with a 4 mm diameter and a length of 17 mm and an inner capsule with a 2 mm diameter and a length of 11 mm. Approximately 30 mg of the BSE mixture was loaded in the small inner capsule and crimped closed. The lower end of the outer capsule was welded,

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