



Conditions of subaerial weathering of basalts in the Neoproterozoic and Paleoproterozoic



Nadezhda A. Alfimova^{a,b,*}, Alexey A. Novoselov^c, Vjacheslav A. Matrenichev^{a,b}, Carlos Roberto de Souza Filho^c

^a Institute of Precambrian Geology and Geochronology, RAS, nab. Makarova 2, St. Petersburg 199034, Russia

^b Saint-Petersburg State University, nab. Universitetskaya 7/9, St. Petersburg 199034, Russia

^c Institute of Geosciences, University of Campinas (UNICAMP), PO Box 6152, Campinas, SP 13083-970, Brazil

ARTICLE INFO

Article history:

Received 28 September 2012

Received in revised form

12 September 2013

Accepted 15 September 2013

Available online 6 October 2013

Keywords:

Chemical weathering

Precambrian

Atmosphere

Climate

Geochemical modeling

ABSTRACT

Precambrian weathering profiles (or regoliths) were formed on the Earth's surface in contact with meteoritic water. This signifies that their mineral and bulk composition should reflect the environmental conditions at the time of weathering. The aim of this study is to reconstruct the main mineralogical and chemical trends involved in the formation of Precambrian weathering regoliths by means of geochemical modeling verified through laboratory experimentation. We have demonstrated that, in a wide range of modeling conditions, weathering profiles of basalts consist mostly of clay minerals (smectites and illite). Carbonate minerals may also be deposited within the profile at the initial stage of weathering, which are later dissolved during a further process. The time, necessary for complete dissolution of primary minerals and crystallization of authigenic minerals is between 100 and 140 Kyr. Modeling results indicate that Neoproterozoic and Paleoproterozoic (2.8–2.0 Ga) weathering profiles were formed under an atmosphere with $p\text{CO}_2$ not much higher than 25 PAL and climatic conditions close to those found in modern times. Weathering under CO_2 -rich ($p\text{CO}_2 = 1$ and 10 bar) and CH_4 -rich atmospheres as well as high temperature (50°C and 75°C) conditions do not produce compounds corresponding to the analyzed samples.

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1. Introduction

Environmental conditions in the Neoproterozoic and Paleoproterozoic are the key for understanding the main trends of climatic changes on a global-scale and they are of vital importance in the point of view of the evolution of life. Throughout that time planetary temperatures varied from below or above the present mean value in the Archean eon to those suitable for global glaciation events in the Paleoproterozoic (Robert and Chaussidon, 2006). The recent revision of oxygen isotopes of mid-Archean cherts (3.5–3.2 Ga), siliceous sediments containing the most continuous record of sea surface temperatures, provides oceanic water temperature estimates no warmer than 40°C (Hren et al., 2009). A subsequent study of ^{18}O in Barberton phosphates rates Archean ocean temperature as high as $26\text{--}35^\circ\text{C}$ (Blake et al., 2010). Dissolution rates of quartz from those cherts are consistent with modern weathering temperatures (Sleep and Hessler, 2006) and thus support conclusion, mentioned above. Nonetheless, accounting for low luminosity of the Sun during the early Precambrian, the increased

pressures of greenhouse gases in the atmosphere seems to be necessary for avoiding the snowball effect (PierreHumbert, 2002). In this context, the carbon dioxide pressure ($p\text{CO}_2$) in the Archean atmosphere could be at least 100–1000 present atmospheric levels (PAL), and perhaps as high as 5–10 bars (Kasting and Ackerman, 1986; Lowe and Tice, 2004). On the other hand, the mineralogy of paleosols and banded iron formations (BIF) suggests much lower CO_2 levels: 7–70 PAL (2.2 Ga, Sheldon, 2006), 3 PAL (Archean, Rosing et al., 2010) and 10–50 PAL (2.69 Ga, Driese et al., 2011), which could not compensate the “faint young Sun” and prevent the surface from freezing. This contradiction can be solved if the atmosphere contained a sufficient amount of other greenhouse gases, such as methane (Lowe and Tice, 2004; Sheldon, 2006; Haqq-Misra et al., 2008; Driese et al., 2011; Zerkle et al., 2012). During the 2.8–2.0 Ga timeframe, planetary scale geologic events took place on Earth, such as an increase in oxygen levels in the atmosphere with a peak at 2.5–2.3 Ga (Rye and Holland, 1998; Holland, 2009; Konhauser et al., 2011) and global glaciations at 2.4–2.3 Ga (e.g., Kasting and Ono, 2006). Precambrian weathering profiles can potentially reflect these features of changing atmospheric compositions and paleoclimates.

Ancient regoliths, formed as a result of chemical weathering under continental subaerial conditions, have a number of distinctions in contrast to modern analogs. Independently of the

* Corresponding author at: Institute of Precambrian Geology and Geochronology, RAS, nab. Makarova 2, St. Petersburg 199034, Russia. Tel.: +7 9213244140.

E-mail address: nadezhda.alfimova@gmail.com (N.A. Alfimova).

substratum composition, their most altered layers are characterized by accumulation of K_2O and Al_2O_3 (Alfimova, 2007). The enrichment with Al_2O_3 due to clay-minerals forming is a trend, typical for the modern hypergen processes as well. However the K_2O content in Proterozoic paleosols is much higher than in modern weathering profiles (e.g. Wiggering and Beukes, 1990). A possible mechanism of K_2O accumulation in the Precambrian is K-metasomatism (Nesbitt and Young, 1989; Maynard, 1992; Fedo et al., 1995) affecting hypergen targets after they were buried. However, the chemical ways of such infiltrations of single element in the system is not clear.

While granitoid series dominated in the composition of Archean continental crust (Martin and Moyen, 2002), mafic rocks were a sufficient component of the dry land surface (Puchtel et al., 1998; Van Kranendonk et al., 2007; Mukasa et al., 2013). According to Taylor and McLennan (1985), the bulk crust composition in the Archean was much more mafic than later on. There are a number of known sites of basaltic regoliths formed due to subaerial weathering in the Neoproterozoic and Paleoproterozoic (Wiggering and Beukes, 1990; Prasad and Roscoe, 1996; Heiskanen and Bondar, 1998; Gorkovetz et al., 1999; Yang et al., 2002; Utsunomia et al., 2003; Alfimova, 2007). Basalts are highly reactive in terms of weathering (Berner and Berner, 1996), and their near surface alteration should have played an important role in sedimentation.

The main goal of this investigation is to thermodynamically and kinetically predict the mineralogical and major-element geochemical changes that occur during continental weathering of basaltic rocks and to obtain constraints on the atmospheric composition and climatic conditions between 2.8 and 2.0 Ga.

2. Geochemical modeling approach

Weathering processes on the Earth's surface occur at low temperature; therefore they are characterized by an incompleteness of chemical reactions. To emulate these low temperature interactions in water–rock systems it is necessary to account for the kinetics of minerals' dissolution. The simulations were implemented with the use of the GEOCHEQ software, which consists of a code for thermodynamic and kinetic calculations and a database developed by Mironenko with co-workers (Mironenko et al., 2008; Mironenko and Zolotov, 2012) on the basis of a thermodynamic database SUPCRT92 (Johnson et al., 1992). The approach used to account for the kinetics of minerals' dissolution was described in Zolotov and Mironenko (2007). This method simulates temporal changes in water–rock systems by coupling pH- and temperature-dependent rate equations of mineral dissolution with calculations of chemical equilibria in aqueous solution. Dissolution of both primary and newly formed secondary phases is assumed to be the rate-limiting process. This approach assumes precipitation is controlled by solubility and occurs faster than mineral dissolution.

2.1. Calculation procedure

Numerical modeling of a weathering profile evolution is implemented by replication of the iterative leaching of rock by aqueous solution with constant composition (depending on the modeling scenario we simulated $n \cdot 10^4 - n \cdot 10^6$ waves). This system is considered open regarding atmospheric gases. The general continuity (t) of the simulated process can be shown as

$$t = w \cdot \sum_{\tau} \Delta t_{\tau}, \quad (1)$$

where w is the quantity of solution waves, $\sum_{\tau} \Delta t_{\tau}$ is the duration of the one solution wave interaction. If $w \rightarrow \infty$, and $\sum_{\tau} \Delta t_{\tau} \rightarrow 0$,¹ the aqueous solution will be strongly unbalanced to bulk volume of rock as well as the substratum will be dissolved effectively.

For the τ th equilibrium computation, the moles $b_{j\tau}$ of a chemical element j in the system are calculated from the chemical composition of aqueous solution at the previous timestep $b_{j(\tau-1)}$ and a sum of molar concentrations of this element $\Delta x_{i\tau}$ from minerals dissolved during the current step:

$$b_{j\tau} = b_{j(\tau-1)} + \sum_i \Delta x_{i\tau} \cdot \nu_{ji}, \quad (2)$$

where ν_{ji} is the stoichiometric co-efficient of a chemical element j in the formula of the i th mineral. The molar amount of the i th mineral dissolved at the τ th timestep is calculated as:

$$\Delta x_{i\tau} = S_{i\tau} \cdot r_{i\tau} \cdot \Delta t_{\tau}, \quad (3)$$

where $r_{i\tau}$ represents the current rate of mineral dissolution (mole $m^{-2} s^{-1}$), $S_{i\tau}$ is the surface area of the i th mineral exposed to solution (m^2), and Δt_{τ} stands for duration of the τ th timestep (s). The initial surface area of the i th mineral can be estimated by means of BET-analysis of specific surface area (S_{SSA}):²

$$S_{i\tau} = \varphi_i \cdot S_{SSA} \sum_l m_l, \quad (4)$$

where φ_i is the volume fraction of the i th mineral in the bulk rock volume, and $\sum_l m_l$ is a weight of all primary or secondary minerals. The dissolution rate ($r_{i\tau}$) equation at timestep τ consists of a Laidler term for the pH-dependence of the dissolution rate, an Arrhenius term for the T-dependence, and an affinity term describing the reduction of the mineral dissolution rate near thermodynamic equilibrium (Zolotov and Mironenko, 2007 and references therein):

$$r_{i\tau} = [k_{OH^+} (a_{H^+})^n + k_{H_2O} + k_{OH^-} (a_{OH^-})^m] \times \exp \left[\frac{E_a}{R} \left(\frac{1}{298.15} - \frac{1}{T} \right) \right] \times \left[1 - \exp \left[\frac{\Delta G}{RT} \right] \right], \quad (5)$$

where k_{OH^+} , k_{H_2O} , k_{OH^-} , n and m are kinetic constants, E_a is the activation energies of mineral dissolution reaction, a_{H^+} and a_{OH^-} are activity of H^+ and OH^- ions, temperature is in Kelvin degrees. The term $\Delta G/RT$ of Lasaga equation may be considered as a measure of saturation. When ΔG (the Gibbs energy change) is close to zero (i.e., near equilibrium), the dissolution rate approaches zero as well. Far from equilibrium, when $\Delta G \ll 0$, the ΔG value has a negligible effect on dissolution rate.

Water–rock ratio (W/R) is to be considered, because it is a proxy for climatic conditions (i.e., rainfall amount). We calculated this as a ratio of rainfall rate to the weight of 1 m rock column (this value is a typical for modern soils): e.g., the rainfall 1000 mm per a year (an average for modern Earth) corresponds to $W/R = 0.036$ for every solution wave. In the algorithm described above, a W/R value used as a reduction factor to calculate $b_{j(\tau=0)}$ (Eq. (2)) from the composition of pure water equilibrated with modeling atmosphere.

¹ In the course of our simulations, the duration of each wave $\sum_{\tau} \Delta t_{\tau}$ was 0.1 year.

² To calculate surface areas of primary minerals we used a SSA of a Proterozoic metabasalt from Northern Karelia (Russia) and for secondary minerals – a SSA of clay, the weathering product of this rock. Both samples were measured by the BET method (Sorbi-M) using nitrogen as an adsorbent gas in the Institute of Structural Microkinetics and Material Science RAS (Chernogolovka, Russia). The SSA of the basalt is $1.432 \pm 0.00547 m^2/g$ and the clay is $53.04 \pm 2.06 m^2/g$.

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