



Gold and silver in a system of sulfide tailings. Part 1: Migration in water flow



I.N. Myagkaya^{a,*}, E.V. Lazareva^a, M.A. Gustaytis^{a,b}, S.M. Zhmodik^{a,b}

^a V.S. Sobolev Institute of Geology and Mineralogy, Siberian Branch of Russian Academy of Sciences, 3, Koptyug Ave. Novosibirsk 630090, Russia

^b Novosibirsk State University, 3, Pirogov Str. Novosibirsk 630090, Russia

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ABSTRACT

Wastes of the ore cyanide process are prone to oxidation and acid leaching that cause mobilization, migration, and precipitation of elements, including noble metals. The issues of waste oxidation, as well as release and transport of oxidation products, are of principal environmental concern and a focus of active geochemical research. We study the behavior of Au and Ag in the natural–industrial system “sulfide wastes–surface/pore waters–particulate matter–bottom sediments” in the presence of different elements (Na, Mg, K, Ca, Al, Fe, Cu, Zn, Se, Ag, Au, Hg, Pb, REE, etc.), at the Novo-Ursk auriferous pyritic deposit (Salair, Kemerovo region, Russia).

The wastes include processed primary ore (wastes I) and ore from the gold-bearing weathering profile (wastes II) that store, respectively, 0.5 ppm Au, 18 ppm Ag and 0.26 ppm Au, 13 ppm Ag. Gold in wastes I occurs in the native form with Cu and Ag impurities and also Au exist as invisible species in pyrite. In wastes II, gold is adsorbed onto the surfaces of secondary mineral particles (kaolinite, montmorillonite, hydromica, and Fe(III) compounds) while silver is an isomorphic impurity in alunite–jarosite minerals. The oxidation of wastes produces acid mine drainage (AMD) water, with pH = 1.9 and high concentrations of sulfate, Fe, Al, Cu, Zn, Pb, As, Se, Te, Hg, Cd, and REE, which flows into the Ur River (a tributary of the Inya).

The stream transports gold and silver existing in dissolved + colloidal and particulate forms. Dissolved + colloidal gold and silver have similar distribution patterns with their contents inversely proportional to pH. Dissolved + colloidal gold in the AMD water is more abundant than silver (0.4–1.2 ppb Au against 0.1–0.3 ppb Ag). Gold changes from the dissolved + colloidal (in AMD) to particulate (till 0.03 ppb) forms and precipitates in progressively larger amounts with distance from the tailings. Silver in most of the analyzed natural and tailings-impacted waters exists as suspended particles (to 1 ppb). The concentrations of gold and silver are the highest (1.8–2.1 ppb Au and 4.5–5.7 ppb Ag) in peat pore waters within the geochemical trains of the tailings, apparently as a result of re-precipitation on organic barriers.

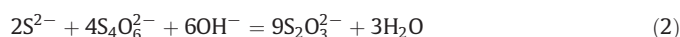
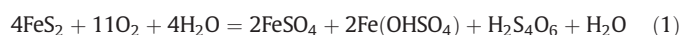
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1. Introduction

Wastes of mining and ore processing have been studied for years in various aspects. The available knowledge on the structure of tailings, as well as on mineralogy, geochemistry, and microbiology of weathering profiles are synthesized in numerous publications (Alpers et al., 1994; Blowes et al., 1994, 2003; Jambor, 1994; Ritchie, 1994; Bigham et al., 1996; Nordstrom, 2000; Dold and Fontbote, 2002; Lazareva et al., 2002; Descostes et al., 2004; Druschel et al., 2004; Gleisner et al., 2006; Equeenuddin et al., 2010; Maescotti et al., 2012; Auld et al., 2013; Sun et al., 2013). This knowledge is especially important as having implications for prevention, treatment, and remediation of contamination effects (Gitari et al., 2008; Ríos et al., 2008; Nyquis and Greger, 2009; Dold et al., 2009; Gibert et al., 2011; Macías et al., 2012; Heviánková et al., 2014; Jeon et al., 2014).

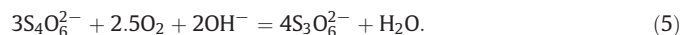
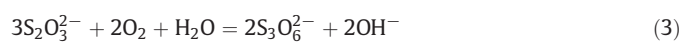
Sulfide wastes deposited in open air are exposed to wind and water weathering and related oxidation, which contaminates large areas

around producing geochemical dispersion haloes and trains (de Haan, 1991; Bortnikova et al., 2003; Moncur et al., 2014). Acid sulfate mine drainage waters formed by oxidation of sulfide materials bear high concentrations of dissolved solids, including heavy metals and toxic elements (Bigham, 1994; Nordstrom and Alpers, 1999; Plumlee, 1999; Seal and Hammarstrom, 2003; Blowes et al., 2003; Valente and Gomes, 2009; Luptakova et al., 2012). Acid mine drainage (AMD) contains abundant sulfate anions and other sulfur ions, such as tetrathionate ($S_4O_6^{2-}$; reaction 1) (Mironov et al., 1989) and thiosulfate ($S_2O_3^{2-}$; reaction 2), according to Pourbaix diagrams (Xia, 2008). Thiosulfate is metastable in the oxide zone and transforms either into tetrathionate ($S_4O_6^{2-}$) or trithionate ($S_3O_6^{2-}$) depending on pH (reactions 3–5); the two latter ions, in turn, become oxidized to SO_3^{2-} , $S_2O_6^{2-}$ and SO_4^{2-} (Xia, 2008).



* Corresponding author.

E-mail address: i_myagkaya@igm.nsc.ru (I.N. Myagkaya).



Elements released from AMD waters become redeposited as ochreous precipitates. Goethite, ferrihydrite, lepidocrocite, jarosite, schwertmannite, and hydrogoethite are main ochreous minerals (Bigham, 1994; Bigham et al., 1996; Maescotti et al., 2012). Iron hydroxides, with their high sorption capacity, commonly adsorb heavy metals and other AMD elements, but ochreous precipitates are unstable and prone to redissolution by acid waters with pH 4.5–6.5 (McDonald et al., 2006; Herrera et al., 2007), which poses pollution problems (Jarvis and Rees, 2004; Carbone et al., 2013). Minor pH-Eh changes can trigger rapid mineral evolution including dissolution, re-precipitation, and structure changes (Maescotti et al., 2012), which return heavy metals back to the waters and maintain widespread of low-density fine-grained ochreous precipitates (Hammarstrom et al., 2003).

The environmental issues concerning acid mine drainage and ochreous precipitates (Hammarstrom et al., 2005; Valente and Gomes, 2009; Sima et al., 2011; Luptakova et al., 2012), and the related contamination (Sarmiento et al., 2011; Carbone et al., 2013), arouse special interest. Most of the research in this line has reasonably focused on the behavior of heavy metals and toxic elements (As, Hg, etc.) (Boulet and Larocque, 1998; Jung, 2001; Lazareva et al., 2002; Al et al., 2006; Roychoudhury and Starke, 2006; Gustaytis et al., 2010, 2013; Equeenuddin et al., 2013; Lusilao-Makiese et al., 2013; Rieuwerts et al., 2014), while noble metals (Au and Ag) in tailings remain little explored (Stoffregen, 1986; Mironov et al., 1989; Benedetti and Boulegue, 1991; Leybourne et al., 2000; Dutova et al., 2006; Reith and McPhail, 2007; Myagkaya et al., 2013).

The behavior of gold in the superficial conditions of oxidation and weathering in gold fields and their surroundings has been mostly studied in terms of distribution patterns, speciation, and morphology of particles (Roslyakov, 1981; Vasconcelos and Kyle, 1991; Lawrance and Griffin, 1994; Bortnikova et al., 1996; de Oliveira and de Oliveira, 2000; Kalinin et al., 2009; Hough et al., 2011; Zhmodik et al., 2012; Reith et al., 2012; Fairbrother et al., 2012). However, some recent papers consider migration paths of gold in mining and refinery wastes (Benedetti and Boulegue, 1991; Leybourne et al., 2000; Al et al., 2006; Roychoudhury and Starke, 2006) and postulate its high mobility in natural and manmade supergene environments (Mann, 1984; Vlassopoulos and Wood, 1990; Vlassopoulos et al., 1990; Andrade et al., 1991; Benedetti and Boulegue, 1991; Howell, 1992; Cidu et al., 1995; Radomskaya et al., 2005; Dutova et al., 2006). Additionally, geochemical cycling of Au in supergene environments can be mediated by microorganisms (Korobushkina and Korobushkin, 1998; Reith et al., 2005, 2012; Reith and McPhail, 2007; Southam and Beveridge, 1994; Southam et al., 2009).

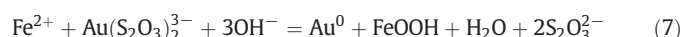
Gold migrating in streams is most often dissolved or particulate. Dissolved gold is present in both natural and mine waters as part of chlorine-, sulfur-, or hydro-complexes, cyanide, as well as mixed Au complexes (Stoffregen, 1986; Vlassopoulos and Wood, 1990; Andrade et al., 1991; Benedetti and Boulegue, 1991; Tossell, 1996; Leybourne et al., 2000; Radomskaya et al., 2005; Dutova et al., 2006; Xia, 2008). As Pourbaix diagrams show, $\text{AuOH}(\text{H}_2\text{O})^0$ (Vlassopoulos and Wood, 1990), and mixed Cl and S complexes such as AuH_2S^+ and AuOHCl^- (Dutova et al., 2006), control gold solubility in most of natural waters (pH to ~6–12). Given the presence of Cl^- , the chloride (AuCl_2^- , AuCl_4^- , AuOHCl^-) and sulfide ($\text{Au}(\text{S}_2\text{O}_3)_2^{3-}$ and $\text{Au}(\text{HS})_2^-$) complexes are the major agents in acid sulfate waters (Mann, 1984; Stoffregen, 1986; Vlassopoulos and Wood, 1990; Andrade et al., 1991; Colin and Vieillard, 1991; Dutova et al., 2006). Thiosulfate Au complexes in acid waters exist according to reaction 6 (Xia, 2008). Furthermore, drainage

solutions may contain cyanide complexes $\text{Au}(\text{CN})_2^-$ that result from oxidative dissolution of wastes (Leybourne et al., 2000).



Particulate gold can form by adsorption onto mineral particles as a result of interaction between hydroxyl groups of colloids and/or cation exchange reactions related to electrostatic interactions. These bonds are relatively weak and the elements are easily desorbed back into water (Sarkar et al., 1999). Second, abundant particulate gold may be due to highly adsorptive amorphous iron hydroxides. Transport of such gold is called mechanic migration (Radomskaya et al., 2005).

Metastable thiosulphate complexes at pH < 5 break down to sulfate, the reaction being catalyzed by transitional elements and bacteria, while gold re-precipitates on iron hydroxide particles (Mironov et al., 1989; Benedetti and Boulegue, 1991; Ran et al., 2002; Yudovich and Ketris, 2004), by reaction 7 (Xia, 2008):



Third, formation of stable colloid particles that can penetrate through a 0.45 μm membrane filter provide the presence of colloids and related elements in filtered water (Petrukhin, 1992; Howe and Clark, 2002; Wang et al., 2003). Fourth, gold can form soluble and insoluble organic complexes which influence its mobility in supergene environments (Baker, 1978; Vlassopoulos et al., 1990; Baranova et al., 1991; Howell et al., 1993a,b; Varshal et al., 1996, 2000; Wood, 1996). For instance, the carboxyl and phenol groups of humic acids (HA) form strong complexes with noble metals, which affects the HA sorption capacity (Varshal et al., 2000) and acts as a barrier for noble metal ions. Interaction with fulvic acids (FA) also leads to the formation of soluble complexes and increases the mobility of noble metals (Baranova et al., 1991; Howell et al., 1993a,b). Furthermore, organic acids can reduce ion complexes and provide gold fixation: e.g., humic substances in river water reduce gold from chloride complexes to gold-bearing organic complexes (Baker, 1978). The standard reduction potentials for FA and HA range from +0.5 to +0.7 V, while the species such as AuCl_4^- and AuCl_2^- are abundant only in highly and moderately oxidizing media, respectively; therefore, humic and fulvic acids do not form dissolved complexes with Au(III) and Au(I) (Wood, 1996). In this case, chloride-complexed gold (AuCl_4^- , AuCl_2^-) becomes reduced and immobilized upon entering the environments where the humic substances have “hard” O-donor groups dominating their binding sites and control the redox conditions. On the other hand, HA cause less influence on reduction and fixation of gold if the Au transporting complexes have low standard reduction potentials (e.g., AuOH^0 ($E^\circ = 0.506$ V), $\text{Au}(\text{HS})_2^-$ and $\text{Au}(\text{S}_2\text{O}_3)_2^{3-}$), with a relatively high proportion of “soft” N- or S-donor binding sites, whereby strong complexing may lower the standard reduction potentials to prevent reduction and promote aqueous transport (Wood, 1996). Organic substances can also maintain reduction of Au (III) to elemental gold and formation of colloids (Ong and Swanson, 1969; Avramenko et al., 2012).

Our previous studies (Myagkaya et al., 2013) showed that oxidative leaching of wastes in the Ursk tailings led to gold and silver dissolution and re-precipitation on a bio-geochemical barrier of peat lying beneath wastes shed from the stockpiles. That inference has guided the research reported in this publication, which consists of two parts. Part 1 deals with Au and Ag migrations in water, in the presence of different elements (Na, Mg, K, Ca, Al, Fe, Cu, Zn, Se, Ag, Au, Hg, Pb, REE etc.) in natural and mine waters in the system “sulfide wastes–surface/pore waters–particulate matter–bottom sediments” in the Ursk area.

2. Study area

The Ursk tailings site is located at 54°27'11.03" N, 85°24'09.76" E in Ursk Village (Kemerovo region, Russia, Fig. 1) within the Ur ore field in

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