



Geochemistry and mineralogy of mine-waste material from a “skarn-type” deposit in central Mexico: Modeling geochemical controls of metals in the surface environment

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ABSTRACT

Several metal sulfide ore deposits (epithermal veins and skarn-type deposits) occur in the central east portion of Mexico (Sierra Madre Oriental). Historically, some of these deposits have been exploited since the 1500s and 1600s. Currently, there are millions of tons of mine waste materials with high content of potentially toxic heavy metals. This work focuses on geochemistry and mineralogy and modeling of redox and acid–base reactions of mine waste material from the Xichú mining area (Aurora Mine), Guanajuato state. The processed ore material at Xichú (circa 1 million tons) consisted of pyrite, arsenopyrite, galena, sphalerite, chalcopyrite, and minerals with Ag and some Au. The mine waste material contains considerable amounts of As, Fe, Pb, Zn, and Cu, in different mineral phases mostly, Fe-oxides and oxyhydroxides, sulfo-salts and traces of primary sulfides. Potential metal liberation and mobility pose a threat to the local population. Redox reactions locally produce pH around 2 in leachates with high metal content. Secondary mineralogy includes goethite, hematite, ferrihydrite, jarosite group minerals, gypsum and minor carbonates, among others. The geochemical dynamics is controlled by the seasonal rains (from June to September), along with high evaporation rates. These factors produce minor acid mine drainage and metal leaching. Redox and dissolution–precipitation reactions control the mobility of heavy metals into the surrounding environment. Arsenic and heavy metal content in tailings is up to 37% Fe, 62 gr/Kg of As, 2.4 gr/Kg of Cu, 13 gr/Kg of Zn and 17 gr/Kg of Pb. Sequential extraction experiments indicated that most of the As is bounded to Fe oxyhydroxides; Zn and Cu are mostly bounded to soluble phases and Pb is mostly bounded to sulfide. Mineralogical changes are controlled seasonally mainly by dissolution and precipitation of secondary phases: jarosite, gypsum, goethite and hematite and minor metal carbonates. Locally, some highly evaporated ponds have low pH (between 2 and 3) and very high concentrations of As and metals. The Geochemist's Workbench (Act2 module) software was used to model the redox conditions for the dissolution–precipitation reaction controlling liberation and mobility of As and metals into the environment. It seems that most of the As and metals are involved in a dissolution–precipitation cycle in the tailings piles and very few are released into the environment.

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1. Introduction

Metals exploitation plays a very important role in the economical, technological and industrial development of the human society. We, as modern human society, are hungry for great amounts of metals to satisfy our needs for modern industry and technology. However, along with the priority for metals exploration comes the basic need for the preservation of a clean environment (sustainable mining development) (Fyfe, 1998; Hill, 1997; Hudson-Edwards et al., 2011; Kesler, 1994; Plant et al., 2001). Regretfully, in most of the historical cases, the exploitation of ore deposits has been

accompanied with a negative impact to the environment (Carrillo et al., 2003; Guogh, 1993; King, 1995; Navarro et al., 2008). It is a fundamental task for all of us to find a balance between exploration and exploitation of metallic ore deposits with the preservation of a clean environment for ourselves and for future generations (Hill, 1997; Hudson-Edwards et al., 2011; Jambor, 1994; Nriagu, 1990; Plant et al., 2001). To face and solve these problems, we need to use all available tools. There is a need for scientific teamwork integrating all the expertise from the natural sciences, the engineering and economic sciences. It is also clear that the activities needed for equilibrated development of ore deposits and the environment are of multidisciplinary character. However, punctual actions, such as environmental geochemistry, mineralogy and geochemical modeling assessments, are basic for future decision-making activities.

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Environmental geochemistry is a relatively new branch of natural sciences (some 30 years) and deals with the analysis of the geologic, mineralogical and chemical processes that introduce and control the concentrations, reactions and mobility of elements or substances potentially contaminant to the environment (Hudson-Edwards et al., 2011; Plant et al., 2001). The distribution of most of these elements or substances could be natural in the environment, but when their concentrations are above certain levels, they become toxic to the ecosystem. The main environmental problems related to ore deposits are acid mine drainage resulting from the oxidation of sulfides (mostly pyrite) and hydrolysis of metal oxides, and high concentrations of heavy metals on water draining from mining sites (Pb, Zn, Cu, Cd, Cr, Fe, Mn, As, Se, Sb, etc.). Since the second half of the 1980s and during the 1990s the environmental research on the impact of mining activities has increased considerably. Some of these research have been focused on the effects of acid mine drainage and high concentrations of heavy metals on surface and groundwater and soil (Alpers and Blowes, 1994; Bowell, 1994; Fyfe, 1998; Haneberg et al., 1993; King, 1995; Larocque and Rasmussen, 1998; Rosner, 1998). Other works have focused on the mineralogical and chemical characterization of metallic ore deposits and their respective wastes (Carrillo and Drever, 1997; Chavez and Walder, 1995; Plumlee and Severson, 1994; Smith et al., 1994). Another research approach has focused on the characterization of mineral surfaces in the natural environment and their effects on dissolved heavy metals (Carrillo and Drever, 1998; Duddley et al., 1988; Koretsky, 2000; Putins and Ruiz-Agudo, 2013; Wuolo, 1986). Still other researches have determined the geochemical baselines to distinguish between natural and anthropogenic sources of metals in the environment (Matschullat et al., 2000; Plant et al., 2001). The use of stable and radiogenic isotopes and recently the use of metal stable isotopes also have found a solid place in environmental geosciences (Bullen and Eisenhauer, 2009; Ghomshei and Allen, 2000; Toner et al., 2003). In addition, environmental geochemical research concerning ore deposits involves geochemical and mathematical modeling, biogeochemistry, molecular environmental geochemistry, laboratory experiments and review on sources, behavior and distribution of individual elements in the near-surface environment (Bener et al., 2000; Blowes et al., 1998; Brown et al., 2000; Early, 1999; Fein et al., 2001; O'Day, 1999; Smedley and Kinniburgh, 2002; Tempel et al., 2000; Zhu and Anderson, 2002). All these geochemical information can be combined with hydrogeological data in order to predict and to model the behavior of heavy metals in groundwater (geometry of plumes, velocity, residence time, etc.). Mine waste materials (mine tailings) account for the world's largest waste material (volumetrically). The geochemical research in all the expertise fields (field and experimental work, analytical, stable isotopes, biogeochemistry and modeling) must increase in order to understand and sustainably manage the historic, contemporary and future mine tailings.

Computational modeling has provided a powerful tool for simulating detailed physical and chemical processes involved in both ore-forming and geo-environmental systems (Gow et al., 2002; Hobbs et al., 2000, 2004, 2007, 2008; Ord et al., 2002; Schaubs and Zhao, 2002; Sorjonen-Ward and Zhang, 2002; Zhang et al., 2003, 2008; Zhao et al., 2008a,b and the references therein included). This enables us to predict the spatial distribution of either ore deposits in an ore-forming system or contaminants in a geo-environmental system with time. For this reason, computational modeling has been widely used to solve various types of geological and geo-environmental problems (Awadh et al., 2013; Charifo et al., 2013; Ju et al., 2011; Khalil et al., 2013; Lin et al., 2003, 2006, 2008, 2009; Liu and Zhao, 2010; Liu et al., 2005, 2008, 2011; Mugler et al., 2012; Xing and Makinouchi, 2008; Yan et al., 2003; Zhao, 2009). In particular, reactive transport modeling, which includes fluid flow, solute diffusion/dispersion and geochemical reaction path, has become an important part of the computational modeling in the field of the emerging computational geoscience (Alt-Epping and Zhao, 2010; Schmidt Mumm et al., 2010; Zhao et al., 2009, 2010). From the scientific point of view, reactive transport modeling should be used

to simulate the spatial distributions of contaminants (such as toxic heavy metals) in geo-environmental systems with time.

In this paper, we present analytical, experimental geochemical data from mine waste materials and some geochemical modeling runs using Geochemist's WorkBench (Bethke, 1996) to assess the reaction paths and mobility of heavy metals in one abandoned mine at the mountainous region of Xichú, Guanajuato State, central-east Mexico. The purposes of this research are:

- (1) to determine the chemical concentrations of heavy metals in the mine waste material;
- (2) to determine the mineralogical phases in which the heavy metals are bound using sequential extractions;
- (3) to use geochemical modeling (Geochemist's WorkBench) to assess the reaction paths and mobility of heavy metals from the mine's waste material to the environment;
- (4) to provide essential information that can be used to construct reactive transport models in the field of the emerging computational geoscience (Zhao et al., 2009).

2. Site description

The Xichú mining district, some 128 Km northeast of Guanajuato City, is located in the core of the natural national preserve of "Sierra Gorda" mountain range (Fig. 1). This mining district is located at the central zone of the folded belt of Jurassic and Cretacic carbonate rocks (Sierra Madre Oriental) of Mexico (Campa and Coney, 1983). Historically, the Aurora mine, located 3 km from the Town of Xichú was exploited since the 1600's, with different periods of exploitation, the last one shutting down operations in 1957. Currently, there are approximately one million ton of mine waste materials with high content of potentially toxic metals. The ore material contains chalcopyrite, galena, sphalerite, and minerals with Ag, Au and As. The mine waste material contains considerable amounts of As, Fe, Cu, Pb, Zn, Cu and Cd posing a threat to the local population. The regional mineralized belt, that includes the Xichú mining district, is related to calc-alkaline igneous intrusions and extensional tectonic stress defined by the NW–SE faulting and fracturing (Randall et al., 1994). At the Aurora Mine, the mineralization is skarn-type with metallic sulfide bodies (Zn, Cu, Pb and Ag). The Ag grade reported was between 350 and 900 gr/tn (COREMI, 1978). The grades reported for other metals were: Zn, 12.12%; Cu, 0.05%; and Pb, 0.45%. The reported ore mineralogy was: pyrite, hematite, galena, sphalerite, and chalcopyrite. The Xichú mines are located at the bottom of the river valley with a marked topographic difference of more than 800 m from the top of the range to the bottom of the valley. More than 1 million ton of metal sulfides-rich mine waste material is located in 4 piles along the Xichú river.

3. Methodology

3.1. Field procedures

The mine waste material is concentrated in four tailing piles along the Xichú River. The tailing piles were labeled tailings 1, tailings 2, tailings 3, and tailings 4 (Fig. 2), from the oldest to the youngest mine waste material. A total of 47 samples were collected from the four tailing piles. The tailing piles have a general stratified structure, with reddish and grayish horizontal layers, and a general hard layer (some 3 cm thick) on the surface composed by Fe oxides and oxyhydroxides protecting sulfide rich material. The samples were collected from a depth of 10 cm (removing the oxidized surface layer) using sterile and previously cleaned plastic ware (hand shovel and bags). Some samples were taken from the same locality and on different layers of the tailing pile (vertical section). Fig. 2 shows the general location of the tailing piles along the Xichú river and the influence zone, the non-influence zone and the mixing zone along the river. In this work, we focus on

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