

Simulating drainage quality of sulfide mine tailings ponds

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Abstract. The Xiangxi gold mine was taken as an example to illustrate the application of the finite element and updated Lagrangian approach in the simulation and prediction of the quality of tailings effluents. The numerical modeling results suggest that tailings-water interaction at the early stage leads to the acidification and release of heavy metals which are responsible for the persistent pollution to the environment for 30 years. Tailings pore water can be gradually neutralized and contents of polluted species tend to decline markedly with time. The gangue dissolution and organic matter reaction can stimulate acid neutralization and decrease the diffusion rate of oxygen in tailings impoundments. The contents of pore-water species in upper tailings are significantly higher than in lower part and change severely at 6 m depth. Therefore, the hydrological zoning phenomenon in tailings impoundments accounts for geochemical zonation. This model has the potential to predict drainage quality of sulfide mine tailing impoundments.

1. Introduction

Tailings–water interaction has been the most important case about water-rock interaction in the surface and kinetic study on this interaction has gained wide attention. Mathematic modeling is a useful tool favorable to understand the physical, chemical and biochemical processes in groundwater systems downgradient of tailings impoundments as well as to learn the complicated interactions among these processes, thus providing many revelations on how to manage tailings impoundments and minimize environment pollution. In our previous investigation, environmental effects of tailings-water interaction of Xiangxi gold mine (XXGM, Fig. 1) tailings, located in western part of Hunan, China, were explored and the results suggest that tailings-water interactions are strong in these areas and result in the production of acid water and release of heavy metals (Au, Sb, As, Hg, Cd, Pb) which contaminated water, soil, plant and other surface environment. In this paper, we conducted a computational numerical simulation on the kinetic process and mechanism of the interaction between XXGM tailings and water based on a coupled process of chemical reaction and mass transport.

2. Computational simulation

2.1. Chemical Mechanism of Tailings–water Interaction

Previous research on mineralogy and geochemistry of XXGM tailings suggested that metallic minerals in tailings comprise sulfide-rich antimonite and pyrite accompanied by minor amounts of



galena, sphalerite, chalcopryrite and arsenopyrite ^[1]. Pyrites contain relatively high content of trace elements, such as Au, Sb, Cu, Pb, Zn, As, Cd. The principal gangue minerals are composed of quartz, calcite, dolomite, chlorite, etc.

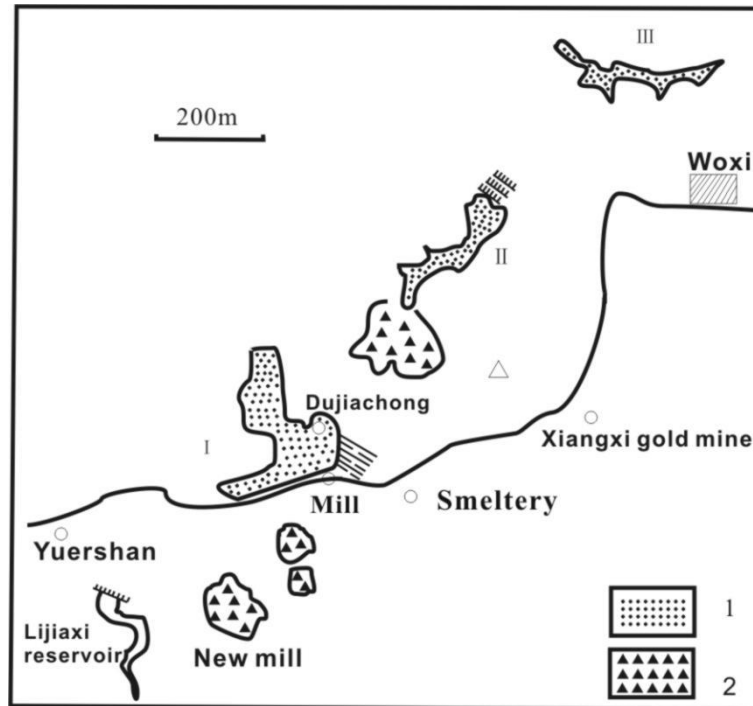
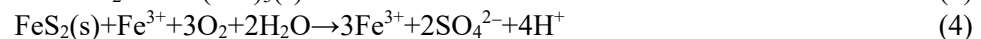
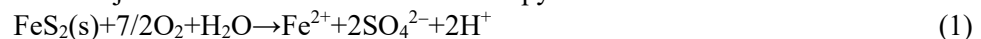
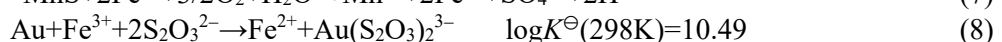
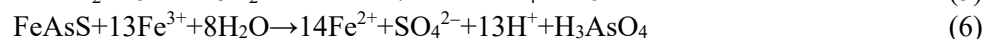


Figure 1. Schematic diagram of Xiangxi gold mine tailings.

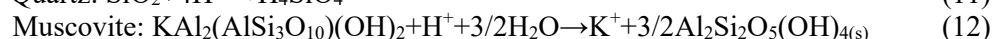
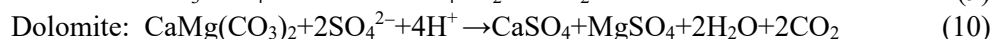
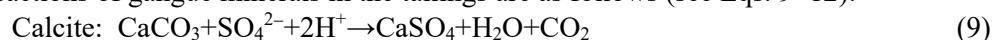
Above all, oxidation of pyrite and other metal sulfides is mainly responsible for the release of acid and heavy metals ^[2,3]. The major chemical reactions involved in pyrite oxidation are as follows:



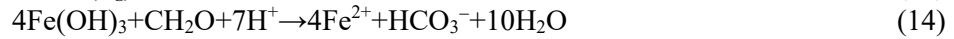
The typical oxidation process of chalcopryrite and arsenopyrite by Fe^{3+} in the surface environment can be described as Eq. 5 and Eq. 6. In addition, oxidation of other metal sulfide (MS) ore intergrowing with pyrite in tailings can be summarized by Eq. 7. It should be noted that significant migration and secondary enrichment of Au element occur in XXGM tailings. Recent research has indicated that thiosulfate as an metastable intermediate produced from sulfide oxidation plays an important role in the dissolution and migration of gold in a surface oxidizing environment ^[4].



Besides the above oxidation reaction of metal sulfide, the dissolution reaction may occur between large amounts of gangue minerals and acid water generated from sulfide oxidation in tailings. The main dissolution reactions of gangue minerals in the tailings are as follows (see Eqs. 9~12):



It should be noted that tailings impoundments, in particular for some old ones whose surface were covered with crops, often contain organic matter. For example, the surface of No.1 and No.2 XXGM tailings impoundments are reclaimed and capped with crops forming organic cover layers. Just due to the organism on tailings surface, the following reactions can also occur:



2.2. Oxygen Transport

As discussed above, oxygen plays an important role in tailing-water interaction, especially in the oxidation process of pyrite and other sulfides. Oxygen can be transported into tailings impoundments by two mechanisms: (1) the infiltration of dissolved oxygen in rainwater into tailings; (2) the diffusion of the gaseous phase oxygen in tailings pore [5]. Assuming that the saturation concentration of dissolved oxygen in rainwater is 8 mg/L at a temperature of 25 °C, if the infiltration rate of rainwater is 0.5 m/y in tailings, then we can deduce that the oxygen flux was 1.3×10^{-10} kg/(m²·s). With the help of stoichiometry coefficient of Eq. 1, we can further work out that the above-mentioned oxygen flux can keep sulfate concentration in the tailings pore about 7 mg/L assuming that all oxygen was reserved in tailing pore with a half amount converted into sulfate. In XXGM, the sulfate concentration in the water affected by tailings is 15.3~52.7 mg/L. Obviously, the dissolved oxygen in rainwater which infiltrated into the tailings plays a great role in sulfide oxidation.

Oxygen is transported through tailings pore mainly by diffusion and an oxygen concentration gradient forms in tailings pore space. The oxygen diffusion coefficient in unconsolidated tailings is related to water content, and the relationship between gaseous diffusion coefficient and air-filled porosity in the unconsolidated sediment can be expressed by the following formula [6]:

$$D_{(\text{O}_2)} = 3.98 \times 10^{-5} [(\phi_g - 0.05) / 0.95]^{1.7} T^{3/2} \quad (17)$$

where $D_{(\text{O}_2)}$ stands for oxygen diffusion coefficient, ϕ_g stands for air-filled porosity, T stands for thermodynamic temperature. Formula (17) shows that oxygen diffusion coefficient in water saturated tailings is far less than in unsaturated ones. When water content in tailings pore is relatively high, it can decrease the air-filled porosity, oxygen diffusion and further prevent sulfide oxidation.

2.3. Mathematical Model

Many research achievements have been made in the dynamic model and computational simulation of water rock interactions [7]. When conducting quantitative simulation of tailings water interaction and production and transport of pollutants, we must take the following physico-chemical processes into account: gas transport and gaseous oxygen concentration in tailings impoundment pore water (TIPW); oxidation reaction of sulfide, dissolution reaction of gangue and other important chemical reactions (organic matter reaction, secondary mineral precipitation, etc); water flow and solute mass transport; heat transport in tailings. The above physico-chemical processes can be described by the a series of equations previous described [1].

3. Simulation results and discussion

According to the previously mentioned model and chemical reaction mechanism of tailings, we designed a computation and modeling program of water-rock interaction in tailings (WRIT) in which different methods were used to solve systems of partial differential equations (all thermodynamic data for various solution and mineral components are provided by EQ3/6). The program can be applied in 1~2dimension numerical simulation in many aspects, such as pH and concentration change of various liquid phase components in TIPW as a function of time and depth of tailings impoundments; the

evolution of the mineral content of various minerals with time and depth of tailings impoundments; the variation of sulfide oxidation rate in tailings with time and depth of tailings impoundments.

In our simulation, the following parameters and initial condition values were chosen based on the geochemical characteristics of XXGM tailings. L is depth of tailings impoundment (20 m); a average grain radius of tailings (0.05 mm); c_w specific heat of water ($4180 \text{ m}^2 \cdot \text{s}^{-1} \cdot \text{K}^{-1}$); $D(\text{O}_2)$ oxygen diffusion coefficient in tailings ($5.0 \times 10^{-6} \text{ m}^2 \cdot \text{s}^{-1}$); ε oxygen consumption per unit mass of sulfur (1.75); ϕ_g air-filled porosity of tailings (0.1); thickness of organic covering layer (0.5 m); ϕ_s total volume fraction of minerals in tailings (0.7); volume fraction of calcite (0.15), pyrite (0.005), arsenopyrite (0.005), galena (0.005), greenockite (0.0001), quartz (0.4108), dolomite (0.1), chalcopyrite (0.005), antimonite (0.008), sphalerite (0.005), gold (0.0001); ρ_s total density of tailings ($1760 \text{ kg} \cdot \text{m}^{-3}$); c_s total specific heat of tailings ($886 \text{ m}^2 \cdot \text{s}^{-1} \cdot \text{K}^{-1}$); ν rainwater infiltration rate in tailings ($0.5 \text{ m} \cdot \text{y}^{-1}$); C_0 oxygen concentration in the air ($0.265 \text{ kg} \cdot \text{m}^{-3}$); δ heat produced per unit mass of sulfide ($2.25 \times 10^7 \text{ J} \cdot \text{kg}^{-1}$); ϕ_w water filled porosity (0.2); organic acid content in the covering layer ($0.05 \text{ kg} \cdot \text{m}^{-3}$).

Fig. 2A was the simulation result of oxygen concentration evolution at the top (1 m depth) and the bottom (19m depth) of the tailings impoundment with time. Fig. 2B reflected the change of oxygen concentration in the tailings impoundment as a function of depth at two different times.

Our simulation results suggested that oxygen concentration at the top and bottom both decreased with time and the former was apparently higher than the latter. Furthermore, a mutation zone existed about 6 m deep. Considering oxygen was the most key factor controlling sulfide oxidation, this process will decrease with time and mainly proceed in the upper part of the tailings impoundment.

The evolution of pH in TIPW with time was shown in Fig. 2C. It appeared that the pH in the upper part of TIPW was approximately 3.0 and possessed acidic property in the first 10 y. During the period of 10~30 y, the pH increased gradually. After 30 y, the pH in the upper part and in the lower part of TIPW both increased progressively and became neutralized. Likewise, the pH of TIPW varied significantly with depth (Fig. 2D). At early stage, the pH in the upper part of TIPW was significantly less than in the lower part with a difference of two units. Similar to the change of oxygen concentration, there was also a pH mutation zone about 6 m deep. With time extended, the pH in the upper part and in the lower part of TIPW both increased toward a constant value 6.0 after 6 y. The No.2 XXGM tailing impoundment was stockpiled about 15 y. Data from field water sample demonstrated that the upper water pH equaled to 4.26, the pH of the water from bottom culvert equaled to 5.33. By contrast, the modeling result showed that the pH at the top (1 m depth) and at the bottom (19 m depth) of TIPW approached 4.13 and 5.51 after 15 y, respectively. Obviously, our simulation results were in primary agreement with the field data.

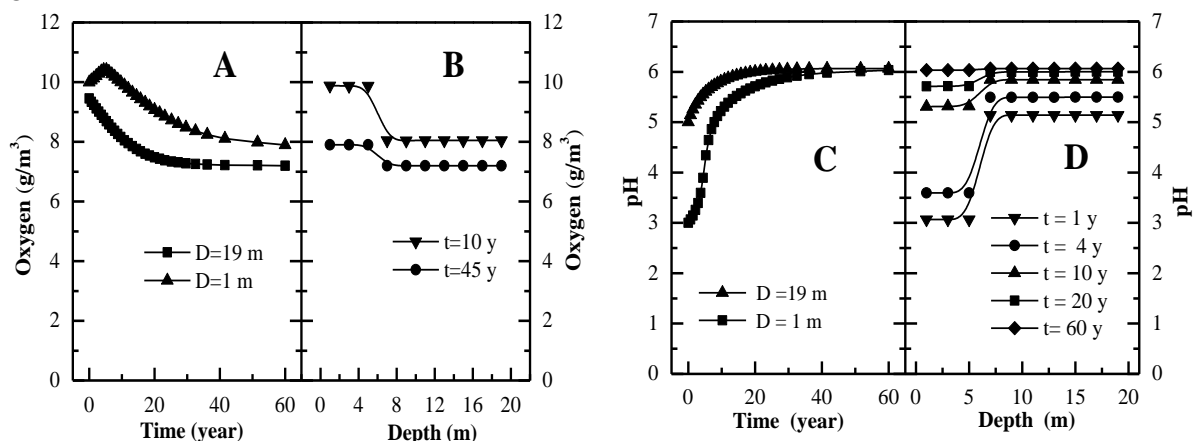


Figure 2. Oxygen levels and pore-water pH in tailing ponds varying with time and depth.

We also simulated the evolution of sulfate concentration [$\sigma(\text{SO}_4^{2-})$] in TIPW with time and depth (Figs. 3A & 3B). As shown in Fig. 3A that sulfate concentration in the upper part and in the lower part of TIPW both increased initially over time and achieved a maximum value within 5~6 y, but reduced subsequently and reached an approximate stable value after about 30 y. Figs. 3A & 3B also reflected that sulfate concentration in the upper part of TIPW were markedly higher than that of in the lower part. Similarly, a mutation zone appeared about 6 m deep. Figs. 2 & 3A also indicated that the pH, oxygen concentration and sulfate concentration of TIPW all reached primary stable values after 30 y, reflecting inherent links and simultaneous evolution of these three variables.

Fig. 3 revealed the simulation result of the variation of Cu, Pb, Zn, Cd, As, Sb content in TIPW with depth after 15 y. Similarly, it is found that the contents of these heavy metal elements were relatively higher in the upper part of PITW than that of in the lower part. Likewise, there was a mutation zone about 6 m deep. In addition, we found that the modeling results was in primary agreement with field data from No.2 XXGM tailings impoundment water body (Table 1). They both suggested that tailings-water interaction resulted in the generation of acid water accompanied with the pollution of Pb, Cd, As, Sb (little pollution of Cu, Zn was detected).

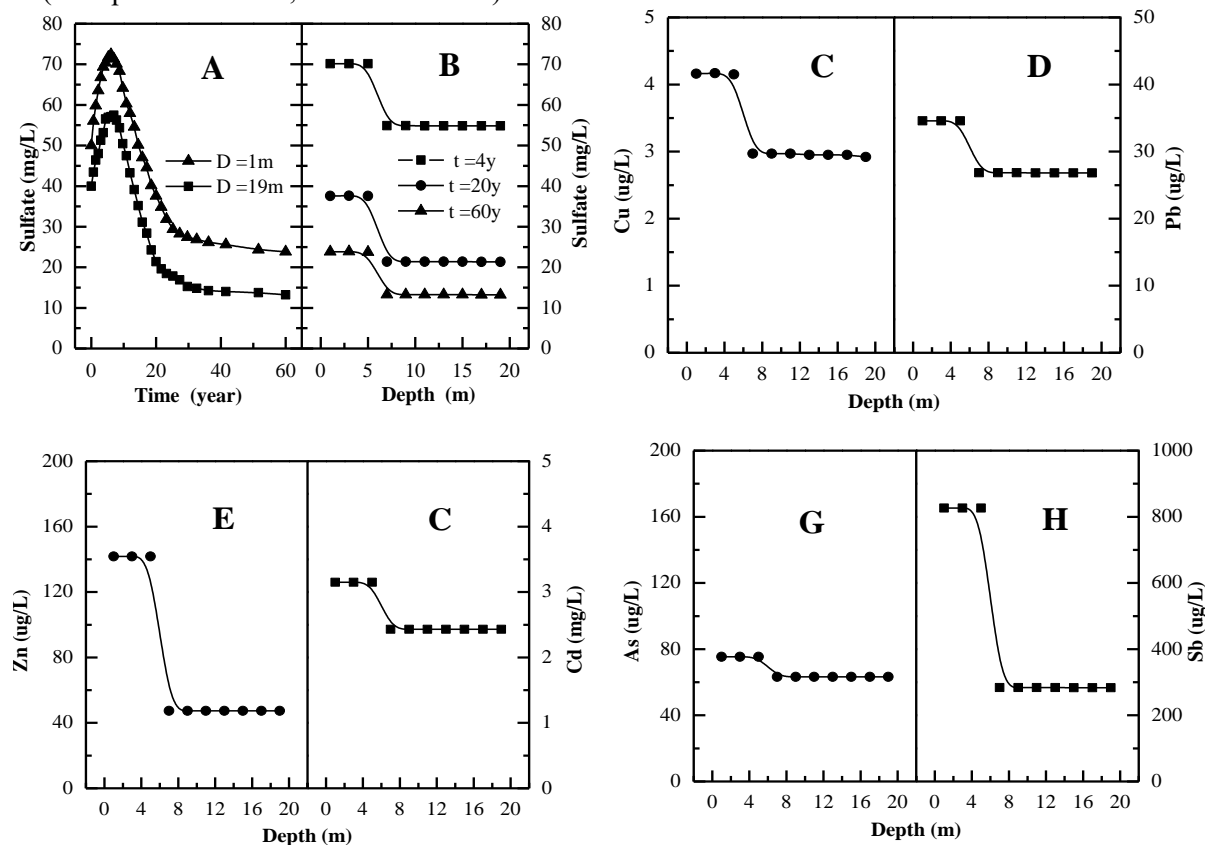


Figure 3. Sulfate and heavy metal levels of pore-water in tailings after 15 y and variation with depth.

The above modeling results demonstrate that the early effect of stockpiling of XXGM tailings impoundments can give rise to the production of acid water and the release of heavy metals which contaminated the environment. The major influence of tailings water interaction on environmental contamination occurred in the first 30 y. As time went on, TIPW gradually tended to be neutralized and pollution element levels decreased significantly. This can be explained by the following reasons. Firstly, tailings became compacted with elapsed stockpiling time of tailings impoundment, thereby making porosity smaller and weakening oxygen diffusion. Meanwhile, the reclamation of tailings impoundment and vegetation growth produced organic matter, the reaction between organic acid and oxygen will consume oxygen in tailings (Eq.13). In the modeling, just considering that the consolidation and

organic acid reaction which can decrease oxygen concentration markedly and further slow the sulfide oxidation (Fig. 2), the production of acid and release of heavy metals declined accordingly. Secondly, the gangue dissolution reaction and organic acid reaction promoted neutralization reaction and sulfate reduction in TIPW, thus leading to the neutralization of water body, solubility decrease of heavy metals and precipitation of metal ore. Finally, as sulfide was gradually oxidized, its content in tailings will decrease and this process will be weakened accordingly.

Our simulation results and field data both indicate that the level of various components in the upper part of TIPW was higher than in the lower part and that a mutation zone existed about 6 m deep. Hydrographic research on tailings impoundment suggested that it can be divided into three hydrographic zones: capillary zone near the groundwater level (central part), rainwater-infiltrated zone (above the capillary zone) and saturated zone (below the capillary zone). Obviously, the air-filled porosity in the upper rainwater-infiltrated zone is larger than that in the lower saturated zone. It can be seen from formula (17) that the diffusion rate and concentration of oxygen in the upper were greater than in the lower saturated part. Modeling results of Fig. 2 also revealed that oxygen level in the upper was markedly greater than in the lower part. Therefore, sulfide oxidation in the upper part was obviously more intensive than in the lower part. Corresponding to hydrographic zonation, the geochemical role of tailings impoundment can be divided into two zones: the upper rainwater-infiltrated zone with sulfide oxidation and acid production; the lower capillary and saturated zone with acid neutralization, sulfate reduction and metal dilution and precipitation. Therefore, the concentration of various components in upper part of the TIPW was obviously higher than in the lower part and a mutation zone appeared near the groundwater surface.

Table 1. Modeling results and experimental values in No.2 XXGM tailings impoundment.

Parameter	Analysis value for No.2 tailings ponds		Simulation results after 15 y	
	Surface	Bottom	Surface	Bottom
$\sigma(\text{Cu})$ [$\mu\text{g/L}$]	3.71	2.73	4.16	2.92
$\sigma(\text{Pb})$ [$\mu\text{g/L}$]	33.7	27.1	34.6	26.8
$\sigma(\text{Zn})$ [$\mu\text{g/L}$]	125.6	22.8	141.7	47.4
$\sigma(\text{Cd})$ [$\mu\text{g/L}$]	2.7	2.2	3.15	2.43
$\sigma(\text{As})$ [$\mu\text{g/L}$]	68.5	59.7	75.4	63.3
$\sigma(\text{Sb})$ [$\mu\text{g/L}$]	733	263	826.5	283.6
$\sigma(\text{SO}_4^{2-})$ [mg/L]	35.7	17.5	47.04	31.12
pH	4.26	5.33	4.13	5.51

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